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Page 119, lines 11-12, after "with", please replace "1-(di-4-n-butylaminophenyl)-10-(4-dimethylaminophenyl)deca 1,3,5,7,9-pentaene" with --1-(di-4-n-butylaminophenyl)-10-(4-dimethylaminophenyl)-deca-1,3,5,7,9-pentane--.

IN THE ABSTRACT:

Please ADD to the specification an ABSTRACT as shown in Appendix E, entitled, "Abstract."

IN THE DRAWINGS:

Please amend the informal drawings as shown in the sheets attached as Appendix F, entitled "Amendments to Drawings."

REMARKS

The Office Action mailed March 11, 2002 required a new preliminary amendment to accomplish the amendments proposed in the preliminary amendment filed July 30, 2001. Entry and consideration of the above proposed amendments and these remarks and re consideration of the subject application are respectfully requested.

Original claims 1, 2, 8-10 and 15 have been cancelled. Claims 3-7, 13 and 14 are pending. Amended claims 3-7, 13 and 14 have been rewritten to be in independent form by including the limitations of the original independent claims from which claims 3-7, 13 and 14 depended. No new matter has been added. Marked up version of amended claims is Appendix A. Clean version of the claims reflecting these amendments accompany this Amendment. Clean version of Amended Claims is Appendix B.

The specification has been amended throughout to clarify the disclosure and improve readability. In addition, the specification has been amended to cross-reference its parent application. No new matter has been added. Applicants note that marked up versions of the paragraphs of the specification in which the above-identified amendments appear are provided in Appendix C, entitled, "Marked Up Version of Amended Paragraphs." Clean versions of the amended paragraphs appear in Appendix D, entitled "Clean Version of Amended Paragraphs."

The subject application has been amended to add an abstract, provided in Appendix E. The specification as a whole supports the abstract; no new matter has been added.

The informal drawings have been amended to clarify the disclosure therein, as indicated in red ink as shown in Appendix F. No new matter has been added.

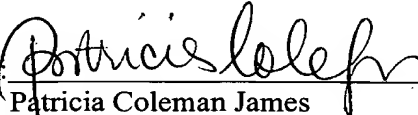
CONCLUSION

In view of the above-made amendments and remarks, Applicants respectfully request an early action on the merits. In the event that a telephonic interview would be helpful for advancing the prosecution, the Examiner is invited to contact the undersigned at (415) 393-2168.

Date: April 24, 2002

Respectfully submitted,

By:



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Appendix D

CLEAN VERSION OF AMENDED PARAGRAPHS

Note: Amended paragraphs are preceded by the location of their first line, indicated in **bold type**.

Page 1, line 16:

B¹ This application is a divisional application of U.S. Application No. 08/965,945 filed November 7, 1997, now U.S. Patent No. 6,267,913 and claims the benefit of U.S. Provisional Application No. 60/029,443 entitled "Two-Photon Optical Limiting Materials," U.S. Provisional Application No. 60/029,437, entitled "Two-Photon Absorbing Polymerization Initiators" and U.S. Provisional Application No. 60/030,141, entitled "Two-Photon Absorbing Optical Materials", all filed November 12, 1996, the disclosure of each of which is incorporated herein by reference.

Page 9, line 13:

B² By the term "chromophore", it is meant a molecule, a molecular fragment, or aggregate of molecules that can absorb electromagnetic radiation.

Page 9, line 25:

B³ By the phrase "optical limiting", it is meant a process wherein the optical transmission of a body decreases with increasing incident optical intensity or fluence such that the intensity or fluence transmitted by the body does not exceed a prescribed level for a prescribed range of incident optical intensity or fluence.

Page 10, line 19:

B⁴ The invention generally provides compositions of matter that have large two-photon or higher-order absorptivities and in some cases large fluorescence quantum yields, leading to efficient two-photon or higher-order excited fluorescence. By two-photon or higher-order absorption, we refer to the initial simultaneous absorption of two or more photons (also referred to as multi-photon absorption), without the actual population of an excited state by the absorption of a single photon. Subsequent to the initial absorption of two or more photons by the molecule, it is possible for the molecule to absorb additional photons from the state populated by the initial absorption event or from a state to which the molecule relaxes.

B4

If sufficient total energy is absorbed by the molecule, it is possible to photoionize the molecule, thereby creating a radical cation of the molecule and an unbound electron. Conversion of a molecule to an excited state by two-photon or higher-order absorption according to the invention enables many applications which can be induced from such an excited state, as described below.

Page 11, line 3:

B5

For example, we have found that stilbene, diphenylpolyene, phenylene vinylene oligomers, and related molecules having two or more electron donor groups, such as amino groups or alkoxy groups, connected to aromatic or heteroaromatic end groups as part of a π (pi)-electron bridge (the combination referred to herein as "D- π -D" molecules) exhibit unexpectedly and unusually high two-photon or higher-order absorptivities in comparison to unsubstituted dyes such as stilbene, diphenylpolyenes, phenylene vinylene oligomers and related molecules, respectively. In addition, the strength and position of the two-photon or higher-order absorption of these molecules can be tuned and further enhanced by appropriate substitution of the π -electron bridge with at least one electron accepting group, such as cyano, (referred to herein as "D-A-D" molecules). We have further discovered that molecules having two or more electron acceptor groups such as formyl or dicyanomethylidene groups, connected to aromatic or heteroaromatic end groups as part of a π (pi)-electron bridge (referred to herein as "A- π -A" molecules) exhibit unexpectedly and unusually high two-photon or higher-order absorptivities in comparison to dyes such as stilbene, diphenylpolyenes, phenylene vinylene oligomers and related molecules. The strength and position of the two-photon or higher-order absorption of these molecules can be tuned and further enhanced by appropriate substitution of the π -electron bridge with electron donating groups, such as methoxy, to form "A-D-A" molecules.

Page 11, line 24:

B6

As a feature of the invention, we have discovered that the aromaticity of the π -bridge in the electronic ground state is important in determining the energetic position of the electronic state which is responsible for multi-photon absorption relative to the state responsible for one-photon absorption. In particular, having donors attached to aromatic groups or heteroaromatic groups, i.e., D- π -D and D-A-D structures, is conducive to the occurrence of the highly absorbing two-photon state energetically above that of the strongly

B₆
fluorescent one-photon state, which is essential for the molecule to exhibit a high multi-photon fluorescence excitation efficiency. In D-A-D molecules, upon absorption of photons, charge is redistributed from the electron donor groups toward the π -electron bridge, thereby enhancing the molecules' photon absorption characteristics.

Page 12, line 6:

B₇
In addition, having acceptors attached to aromatic groups or heteroaromatic groups, i.e., A- π -A and A-D-A structures, is conducive to the occurrence of the highly absorbing two-photon state energetically above that of the strongly fluorescent one-photon state. In A-D-A molecules, upon absorption of photons, charge is transferred from the π -electron bridge toward the electron accepting groups, thereby enhancing the molecules' photon absorption characteristics.

Page 12, line 12:

B₈
Accordingly, the combination of the intrinsically high fluorescence quantum efficiency for these molecules and the rapid relaxation of the two-photon excited molecule from the two-photon state to the lower lying fluorescent one-photon state makes the molecules according to the invention highly efficient multi-photon excitable fluorescent emitters.

Page 14, line 21:

B₉
These new two-photon or multi-photon absorbers may be used for two-photon or multi-photon two-dimensional or three-dimensional optical lithography and three dimensional optical memory.

Page 15, line 19:

B₁₀
Upon multi-photon excitation, these molecules become very strong excited state reducing agents and can therefore be used as multi-photon excited electron donors. Such materials can transfer electrons to electron acceptors (which may be covalently attached to the multi-photon absorbing chromophore or could be in a region of space near the multi-photon absorbing chromophore, as would be the case in a concentrated fluid solution or solid solution) to create a radical cation, in the case of the multi-photon absorbing dye and a radical anion, in the case of the acceptor.

Page 15, line 27:

B¹²

Another feature of some molecules in one embodiment of the invention is that molecules "end capped" with electron accepting substituents, i.e., A- π -A and A-D-A structures, have relatively high electron affinities in the ground-state form. Furthermore, upon multi-photon excitation, these molecules become very strong excited state oxidizing agents and can therefore be used as multi-photon excited electron acceptors. Such materials can accept electrons from electron donors (which may be covalently attached to the multi-photon absorbing chromophore or could be in a region of space near the multi-photon absorbing chromophore as would be the case in a concentrated solution or solid solution) to create a radical anion, in the case of the multi-photon absorbing dye, and a radical cation, in the case of the donor.

Page 16, line 11:

B¹³

In addition, these radical ion species generated upon multi-photon absorption can be chemical reactants. For example, these multi-photon absorbing materials can be used to initiate polymerization of monomers which can be polymerized either by radical or ionic mechanisms in the presence or absence of polymerization promoters, including chain transfer agents. Examples of monomers which can be polymerized in these ways include, but are not limited to, vinyl monomers including acrylonitriles, acrylates, diacrylates, triacrylates, polyacrylates, styrenes, trifunctional isocyanurates substituted with acrylates, vinyl- and allyl-containing monomers, divinylbenzene and polymers which are end-capped with or have these groups attached to the carbon backbone such as polyurethanes, polyesters and polyolefins. Additional examples of polymerizable monomers may be found in G. Odian, *Principles of Polymerization*, Second Edition, John Wiley and Son, New York, 1981, page 181, which is incorporated herein by reference.

Page 18, line 1:

B¹⁴

Another feature of this invention is that it is possible to tune the lipophilicity, hydrophilicity and overall solubility of the multi-photon absorbing chromophores over a very wide range by the appropriate substitution of the donor groups. For example, 4,4'-bis-dimethylaminostilbene and 4,4'-bis-(diphenyl)aminostilbene are sparingly soluble in nonpolar organic solvents whereas 4,4'-bis-(di-*n*-butylamino)stilbene and 4,4'-bis(di-(4-*n*-butylphenyl)aminostilbene are very soluble, i.e., are lipophilic. In contrast, the hydrochloric acid

B14
adduct of the bis-lysyl ester of 4-diethylamino 4'-diethanolaminostilbene and the bis-lysyl ester of 1-(4-dimethylaminophenyl)-4-(4'-diethanolaminophenyl) buta-1,3-diene shown below are hydrophilic. In each case, the molecules maintain their fluorescence in organic and aqueous solution.

Page 19, line 28:

B15
An advantage which is gained by the efficient multi-photon absorption processes enabled by molecules according to the invention is that it is possible for the absorption to occur in a region of space whose cross sectional area is smaller than that of the diffraction-limited spot size of the exciting beam of light. This is because, for example, a two-photon process is dependent on I^2 , where I is the intensity of the exciting beam. The diffraction-limited spot size for a Gaussian beam is given by:

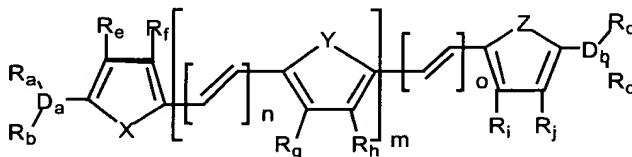
$$d_{1/e^2} = \frac{0.635\lambda}{NA}$$

where λ is the wavelength of the exciting beam of light and NA is the numerical aperture of the focusing lens, and d_{1/e^2} is the full width of the beam where the intensity is $1/e^2$ times the peak intensity. Therefore the theoretical spot size for a two-photon excitation is given by:

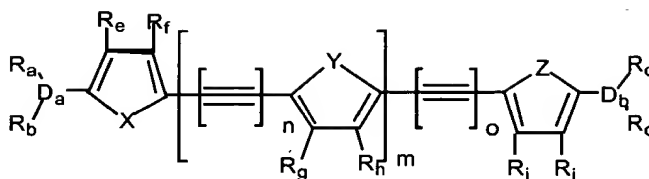
$$d_{\text{"2-photon"}} = \frac{d_{1/e^2}}{\sqrt{2}}$$

Page 21, line 6:

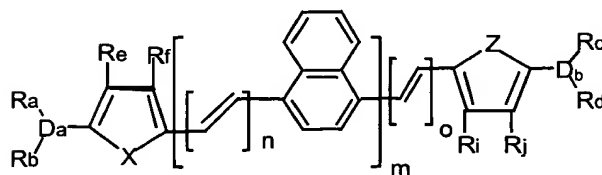
Class 1 Structures: Compounds where the end groups are electron donating groups



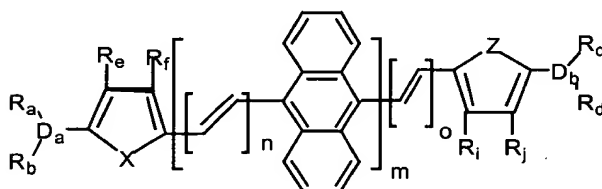
Class 1- I



Class 1- II



Class 1- III



Class 1- IV

where D_a is any electron donating group, that is any one of N, O, S and P;

where D_b is any electron donating group, that is any one of N, O, S and P;

m, n, o are integers such that $0 \leq m \leq 10$, $0 \leq n \leq 10$, $0 \leq o \leq 10$; and

where X, Y, Z are independently selected from: $CR_k=CR_l$; O; S; $N-R_m$

where $R_e, R_f, R_g, R_h, R_i, R_j, R_k, R_l, R_m$ are defined in NOTE 3.

Page 22, line 8:

R_a, R_b, R_c, R_d are independently selected from: H; a linear or branched alkyl group with up to 25 carbons; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta CO_2 R_{\alpha 1}$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta OR_{\alpha 1}$;

$-(CH_2CH_2O)_\alpha-(CH_2)_\beta NR_{\alpha 2} R_{\alpha 3}$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta CONR_{\alpha 2} R_{\alpha 3}$;

$-(CH_2CH_2O)_\alpha-(CH_2)_\beta CN$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta Cl$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta Br$;

$-(CH_2CH_2O)_\alpha-(CH_2)_\beta I$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta$ -Phenyl; various aryl groups (see NOTE 1);

various fused aromatic rings (see NOTE 2); various polymerizable functionalities attached through a linkage which can be chosen from a linear or branched alkyl chain with up to 25

carbons, various aryl groups, $(\text{CH}_2\text{CH}_2\text{O})_\alpha\text{-(CH}_2)_\beta\text{-}$, and $\text{-(CH}_2\text{CH}_2\text{O})_\alpha\text{-(CH}_2)_\beta\text{CO}_2$ (see NOTE 2A);

- B¹⁷
- R_{a1} , R_{a2} , and R_{a3} are independently selected from: H; a linear or branched alkyl group with up to 25 carbons;
 - R_{a1} , R_{a2} , and R_{a3} can also be a functional group derived essentially from one of the 20 naturally occurring amino acids: alanine; valine; leucine; isoleucine; proline; tryptophan; phenylalanine; methionine; glycine; serine; threonine; tyrosine; cysteine; glutamine; asparagine; lysine; arginine; histidine; aspartic acid; and glutamic acid; a polypeptide; adenine; guanine; tyrosine; cytosine; uracil; biotin; ferrocene, ruthenocene, cyanuric chloride and derivatives thereof, or methacryloyl chloride. By "derived essentially", it is meant that R_{a1} , R_{a2} , and R_{a3} may be an amino acid or a functional group that is closely related (e.g., a trivial modification of an amino acid).
 - where α is 0-10,
 - where β is 1-25.

Page 24, line 18:

B⁸

R_e , R_f , R_g , R_h , R_i , R_j , R_k , R_l , R_m are independently selected from: H; a linear or branched alkyl group with up to 25 carbons; $(\text{CH}_2\text{CH}_2\text{O})_\alpha\text{-(CH}_2)_\beta\text{CO}_2\text{R}_{a1}$; -
 $(\text{CH}_2\text{CH}_2\text{O})_\alpha\text{-(CH}_2)_\beta\text{OR}_{b1}$;
 $\text{-(CH}_2\text{CH}_2\text{O})_\alpha\text{-(CH}_2)_\beta\text{NR}_{b2}\text{R}_{b3}$; $\text{-(CH}_2\text{CH}_2\text{O})_\alpha\text{-(CH}_2)_\beta\text{CONR}_{b2}\text{R}_{b3}$;
 $\text{-(CH}_2\text{CH}_2\text{O})_\alpha\text{-(CH}_2)_\beta\text{CN}$; $\text{-(CH}_2\text{CH}_2\text{O})_\alpha\text{-(CH}_2)_\beta\text{Cl}$; $\text{-(CH}_2\text{CH}_2\text{O})_\alpha\text{-(CH}_2)_\beta\text{Br}$;
 $\text{-(CH}_2\text{CH}_2\text{O})_\alpha\text{-(CH}_2)_\beta\text{I}$; $\text{-(CH}_2\text{CH}_2\text{O})_\alpha\text{-(CH}_2)_\beta\text{-Phenyl}$; various aryl groups (see NOTE 1); various fused aromatic rings (see NOTE 2); various polymerizable functionalities (see NOTE 2A); $\text{NR}_{e1}\text{R}_{e2}$; OR_{e3} ; CHO; CN; NO₂; Br; Cl; I; phenyl; an acceptor group containing more than two carbon atoms (see NOTE 3A), and where R_{e1} , R_{e2} , R_{e3} , are independently defined as for R_n and R_o in NOTE 3B;

- R_{b1} , R_{b2} , and R_{b3} can also be a functional group derived essentially from one of the 20 naturally occurring amino acids: alanine; valine; leucine; isoleucine; proline; tryptophan; phenylalanine; methionine; glycine; serine; threonine; tyrosine; cysteine; glutamine; asparagine; lysine; arginine; histidine; aspartic acid; and glutamic acid; or a polypeptide; adenine; guanine; tyrosine; cytosine; uracil; biotin; ferrocene; ruthenocene, cyanuric chloride and derivatives thereof or methacryloyl chloride. By "derived essentially from", it is meant that R_{b1} , R_{b2} , and R_{b3} can be an amino acid or a functional group that is closely related to an amino acid (i.e., a trivial modification of an amino acid).

B¹⁸

- where α is 0-10,
- where β is 1-25.

Page 27, line 5:

R_n , R_o are independently selected from: H; a linear or branched alkyl group with up to 25 carbons; $(CH_2CH_2O)_\alpha-(CH_2)_\beta CO_2R_{\alpha 1}$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta OR_{g1}$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta NR_{g2}R_{g3}$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta CONR_{g2}R_{g3}$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta CN$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta Cl$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta Br$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta I$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta$ -Phenyl; various aryl groups (see NOTE 1); various fused aromatic rings (see NOTE 2); various polymerizable functionalities (see NOTE 2A);

R_{g1} , R_{g2} , and R_{g3} are independently selected from: H; a linear or branched alkyl group with up to 25 carbons;

B¹⁹

- R_{g1} , R_{g2} , and R_{g3} can also be a functional group derived essentially from one of the 20 naturally occurring amino acids: alanine; valine; leucine; isoleucine; proline; tryptophan; phenylalanine; methionine; glycine; serine; threonine; tyrosine; cysteine; glutamine; asparagine; lysine; arginine; histidine; aspartic acid; and glutamic acid; or a polypeptide; adenine; guanine; tyrosine; cytosine; uracil; biotin; ferrocene, ruthenocene, cyanuric chloride and derivatives thereof or methacryloyl chloride. By "derived essentially from" it is meant that R_{g1} , R_{g2} , and R_{g3} can be an amino acid or a functional group that is closely related to an amino acid (i.e., a trivial modification of an amino acid).
- where α is 0-10, and where β is 1-25.

Page 31, line 7:

R_a , R_b , R_c , R_d are independently selected from: H; a linear or branched alkyl group with up to 25 carbons; $(CH_2CH_2O)_\alpha-(CH_2)_\beta CO_2R_{\alpha 1}$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta OR_{a1}$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta NR_{a2}R_{a3}$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta CONR_{a2}R_{a3}$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta CN$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta Cl$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta Br$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta I$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta$ -Phenyl; various aryl groups (see NOTE 1 as described above for Class 1 molecules); various fused aromatic rings (see NOTE 2 as described above for Class 1 molecules); various polymerizable functionalities (see NOTE 2A

B²⁰

B²⁰
as described above for Class 1 molecules); NR_{e1}Re₂; OR_{e3}; CN; NO₂; Br; Cl; I; phenyl;
or an acceptor group containing more than two carbon atoms (see NOTE 3A as described
above for Class 1 molecules).

Page 32, line 3:

B²¹
The compositions of the invention are organic molecules having multi-photon
absorption properties. The compositions themselves may exist as crystals, mesoscopic
phases, polymers, glasses, liquids or gases. The compositions may be used alone or in
combination with other crystals, mesoscopic phases, polymers, glasses liquids or gases.

Page 33, line 3:

B²²
Generally, the methods according to invention are carried out by converting a multi-
photon absorbing molecule to an electronically excited state by absorption of at least two
photons of radiation. The excited state then facilitates numerous applications. The molecule
may be irradiated with visible, ultraviolet or infrared radiation to effect the multiphoton
absorption.

Page 54, line 3:

B²³
0.01 mol of the appropriate dialcohol, 0.03 mol of the amino acid and 0.025 mol of
hydroxybenztriazole (HOBT·H₂O) were dissolved in 60 mL of anhydrous N,N-
dimethylformamide and stirred under nitrogen atmosphere. After cooling down to 0°C, 0.035
mol of dicyclohexylcarbodiimide were added at once. The solution was allowed to warm to
ambient temperature and stirred for further 15 h. The precipitated urea was filtered off and
the solution was diluted with 200 mL of diethyl ether. The organic phase was washed three
times with 50 mL of water, dried over Na₂SO₄ and evaporated to dryness. To remove the
excess of amino acid, the residue was dissolved in diethyl ether and filtered over a short
column of Florisil. The organic solvent was evaporated again yielding the bisester as a solid.
Yields were typically around 70-80%.

Page 72, line 7:

B24
1,4-Bis(4-morpholino)styryl Benzene : prepared from the reaction of 4-morpholinobenzaldehyde (3.94 g, 0.02 mol) and 1,4-bis(methyltriphenylphosphonium chloride) (8.42g, 0.012 mol) and lithium/ethanol (0.296g, 0.043 mol/ 150 mL).

Recrystallized from ethanol to yield a mixture of isomeric products, Yield 8%;

Recrystallized from acetone to yield one pure isomer, Yield 11.8%; m.p. = 155°C; IR (KBr) ν cm^{-1} : 1604, 1519, 1355, 1179, 962, 822; UV-Vis (acetone) λ_{max} , nm: 400; ^1H NMR (CDCl_3 , δ): 3.66 (t, J = 6.9 Hz, 8H), 3.76 (t, J = 6.9 Hz, 8H), 6.68, 7.42 (d, J = 8.5 Hz, 4H), 7.03 (d, J = 16.30 Hz, 2H), 6.96, (d, J = 16.30 Hz, 2H), 7.42 (s, 4H); MS (FAB, m-NBA matrix): m/z 560.92 (M^+); MS (EI): m/z 561.9, 512.9, 462.0, 436.0, 399.0, 336.0.

Page 72, line 20:

B25
1,4-Bis(4-di-n-butylamino)styryl Anthracene: prepared from the reaction of 9,10-anthracenedicarboxaldehyde (1.17 g, 0.005 mol) and 4-dibutylaminobenzyl phosphonium iodide (7.563g, 0.0125 mol) and lithium/ethanol (0.178g, 0.0295 mol/ 70 mL).

Recrystallized from N,N-dimethylformamide/water, Yield 62.4%; Anal. Calcd. for $\text{C}_{46}\text{H}_{56}\text{N}_2$: C, 86.73; H, 8.86; N, 4.39. found: C, 86.18; H, 8.59; N, 5.35; IR (KBr) ν cm^{-1} : 2948, 1607, 1518, 1365, 1186, 760; UV-Vis (acetone) λ_{max} , nm: 440, 320; ^1H NMR (C_6D_6) δ : 0.870 (t, J = 7.2 Hz, 12H), 1.18 (m, 8H), 1.48 (m, 8H), 3.08 (t, J = 7.4 Hz, 8H), 6.71, 7.51 (Ar, J = 8.5 Hz, 8H), 6.98 (d, J = 16.4 Hz, 2H), 7.82 (d, J = 16.4 Hz, 2H), 7.34, 8.66 (m, 8H, Ar); MS (FAB, m-NBA matrix): m/z 636.4 (M^+).

Page 76, line 19:

B26
The general procedure for the double Wittig reaction using NaOEt was followed. After 4h of reflux, the reaction mixture was cooled and water was added. Upon addition of water, an oily product formed that became a solid mass after cooling overnight. The yellow solid was obtained in 94.9% yield as a mixture of isomers. (In the above structure, the "sawtooth" bond indicates that the structure includes both cis and trans isomers.)

Page 79, line 16:

B27
Figure 10 illustrates the reaction of 1,4-bis(cyanomethyl)benzene with 4-diphenylaminostilbene and 4-di-n-butylaminobenzaldehyde to yield 1,4-bis(β -cyano-4'-diphenylaminostyryl)benzene and 1,4-bis(β -cyano-4'-di-n-butylaminostyryl)benzene with donor side units and acceptors on the vinylene units (see examples 72 and 73).

Page 90, line 2:

B28

For comparison with the data in Table 2, *para*-bis(*ortho*-methylstyryl)benzene, which lacks donor or acceptor substitution, exhibits a two-photon absorptivity of $55 \times 10^{-50} \text{ cm}^4 \text{ s/photon}$ at 568 nm [C. Xu and W. W. Webb, J. Opt. Soc. Am. B, **13**, 481 (1996) and S. M. Kennedy and F. E. Lytle, Anal. Chem. **58**, 2643 (1986)]. In accord with the present invention compounds symmetrically substituted with a pair of terminal donors, [i.e., D- π -D] or symmetrically substituted to form molecules according to the donor-acceptor-donor (D-A-D) or acceptor-donor-acceptor (A-D-A) motifs exhibit substantially enhanced two-photon absorptivities compared to unsubstituted *para*-bis(*ortho*-methylstyryl)benzene.

Page 90, line 12:

B29

Measurements of two-photon absorptivity by use of nonlinear optical transmission method in solutions of bis-donor-substituted diphenylpolyenes show a strong dependence on laser pulse duration, as indicated by the values in Table 3. These results provide evidence of nonlinear absorption that is higher-order than two-photon absorption. In particular, the results suggest that these molecules undergo two-photon induced excited state absorption, that is, two-photon absorption to an excited state and further absorption from this excited state or from a state formed rapidly on relaxation from the initial excited state. A greater excited state population is created with the longer pulses, leading to the enhanced nonlinear absorption coefficient. Additional measurements including transient photoinduced absorption measurements and picosecond time resolved pump and probe measurements confirm the presence of excited state absorption induced by two-photon absorption, which occurs at the two-photon excitation wavelength. The effective two-photon absorptivities of the series of bis-donor diphenylpolyenes is one to two orders of magnitude larger than that measured with picosecond pulses and, in accord with the present invention, these data demonstrate utility in optical limiting applications involving nanosecond laser pulses. Furthermore, the values of the effective two-photon absorptivity obtained for the bis donor diphenylpolyenes are an order of magnitude larger than that of Rhodamine B.

Page 91, line 12:

B30

Table 3. Effective two-photon absorption data on a series of bis-donor diphenylpolyenes as measured with nanosecond (ns) and picosecond (ps) pulses and

B³⁰ nonlinear optical transmission methods. The units of the two-photon absorptivity, δ , are $10^{-50} \text{ cm}^4\text{-s/photon}$.

Page 98, line 3:

B³¹ Here we describe the utility of two-photon and multi-photon absorbing molecules as initiators of polymerization, specifically to the use of bis-donor-substituted π -conjugated molecules as two-photon initiators. Such two-photon excitable initiators have utility in two- and three-dimensional lithography applications, and in three-dimensional optical data storage with ultrahigh storage densities ($10^{12} \text{ bits/cm}^3$). When applied to a spinning disk (compact disk) type format, two-photon polymerizable materials can offer over a thousand-fold increase in storage capacity. There are two key advantages of two-photon induced polymerization relative to single-photon induced polymerization. Whereas single-photon absorption scales linearly with the intensity of the incident radiation, two-photon absorption scales quadratically. As a result, it is possible to perform two-photon polymerization with three-dimensional spatial resolution. In addition, because the phenomenon involves the simultaneous absorption of two-photons, the chromophore is excited with light of half the energy of the two-photon absorption peak. As a result, it is possible to excite molecules at a greater depth within a material than would not be possible via single-photon excitation. These two advantages also apply to for example *in vivo* photo-initiated polymerization. In lithography or stereolithography, the quadratic scaling of absorption with intensity can lead to the ability to write features below the diffraction limit of light and the ability to write features in three dimensions, which is also of interest for holography.

Page 110, line 10:

B³² A critical issue related to *in vivo* photopolymerization addressed in this invention is limitations that may be introduced by the use of visible light; specifically, visible light does not penetrate tissues or other absorbing or scattering media at depth. To address this deficiency, we have developed photoinitiators which can be activated by two-photon absorption of near-infrared light, which has the greatest penetration depth through tissue.

Page 115, line 26:

B³³ The new multi-photon absorbers according to the present invention afford several interesting advantages for optical limiting. Firstly, these molecules exhibit effective two-

B33
photon absorptivities that are about one or more orders of magnitude higher for current state of the art materials. With such high two-photon absorptivities, these molecules are effective in limiting nanosecond duration pulses. Secondly, these molecules can have very high broadband linear transmission, since the two-photon absorption bands occur for photon energies well below the linear absorption edge. This could be a huge advantage, since low linear transmission and impairment of normal color vision are major concerns for potential users of laser protective devices. Thirdly, broadband two-photon absorbing molecules have been prepared from mixtures of compounds, all of which are essentially transparent over much of the visible spectrum, each with different two-photon resonances that span a wide band. Finally, these two-photon absorbing molecules perform very well at high intensities such that good short pulse performance is assured.

Page 116, line 12:

B34
We have demonstrated optical limiting based on the new molecular two-photon and multi-photon absorbers described herein in different optical geometries and with combinations of materials as described below. The optical limiting compositions according to the present invention may exist as crystals, mesoscopic phases, polymers, glasses, liquids or gases. The compositions may be used alone or in combination with other crystals, mesoscopic phases, polymers, glasses, liquids or gases. By "mesoscopic phases," we refer to materials with structural order on a length scale between that of individual molecules, i.e., above about 10 Angstroms, and the microscopic length scale, i.e., above about one micrometer. These materials include small molecule and polymeric liquid crystals, colloidal, micellar and liposomal suspensions, self assembled nanoparticle arrays, and the like.

Page 119, line 3:

B35
A mechanism to enhance the two-photon absorptivity at a given wavelength is to synthesize molecules with extended conjugation such that the single-photon absorption edge approaches the fundamental frequency of the laser, without leading to substantially decreasing linear loss. For example, simply extending the number of bridging double bonds in the stilbene derivatives from one to five double bonds enhances the effective two-photon absorptivity by a factor of five. The enhanced two-photon absorption is evidenced in Figure 19 where the nonlinear transmission of 4,4'-bis-(di-n-butylamino)stilbene is compared with 1-(di-4-n-butylaminophenyl)-10-(4-dimethylaminophenyl) deca-1, 3, 5, 7, 9-pentene.



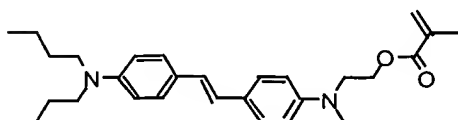
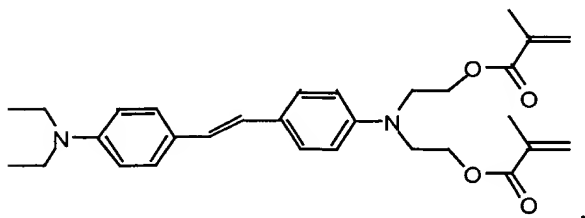
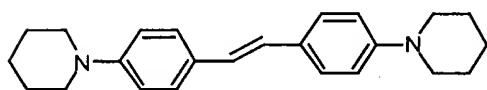
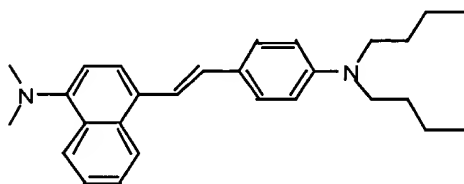
Appendix A

MARKED UP SET OF AMENDED CLAIMS

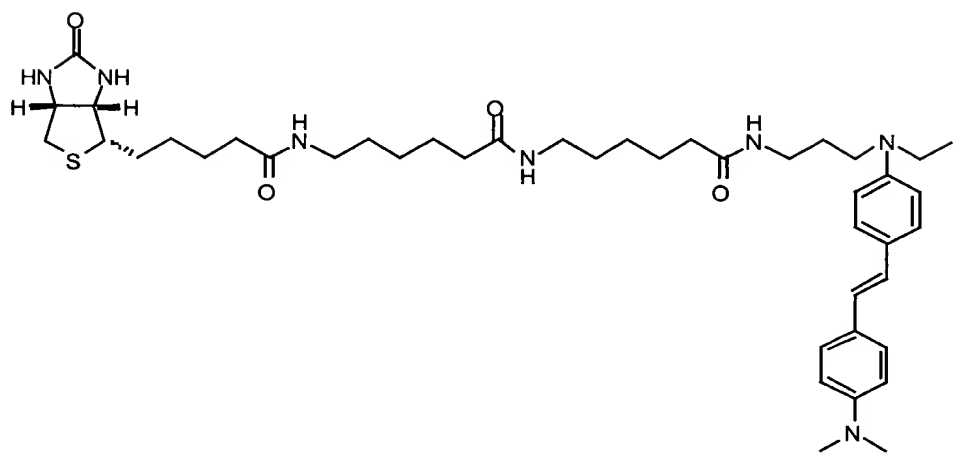
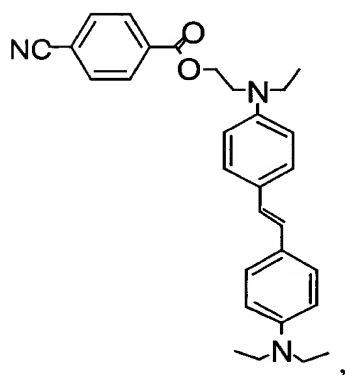
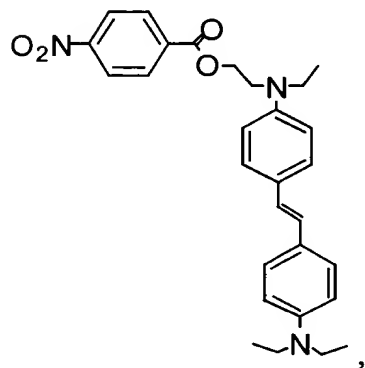
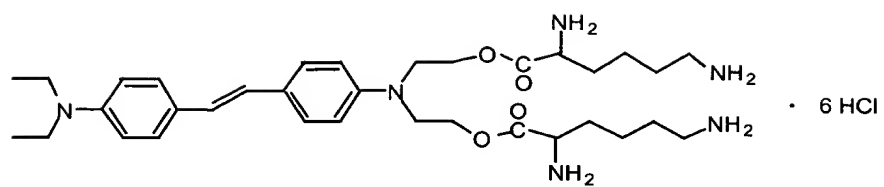
3. (Amended) A method for preparing a compound in an electronically excited state, comprising the steps of:

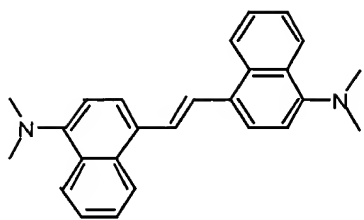
a) exposing a compound having the formula D_1 - Π - D_2 to radiation, wherein D_1 and D_2 are electron donor groups; and Π comprises a bridge of π -conjugated bonds connecting D_1 and D_2 ; and

b) converting said compound to a multi-photon electronically excited state upon simultaneous absorption of at least two photons of said radiation by said compound, wherein the sum of the energies of all of said absorbed photons is greater than or equal to the transition energy from a ground state of said compound to said multi-photon excited state and wherein the energy of each absorbed photon is less than the transition energy between said ground state and the lowest single-photon excited state of said compound and is less than the transition energy between said multi-photon excited state and said ground state [A method according to claim 2], wherein said compound is selected from the group consisting of

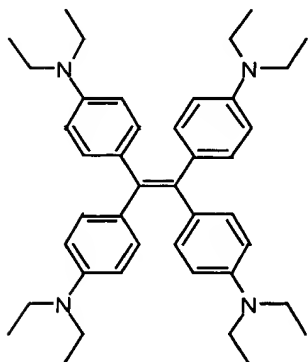


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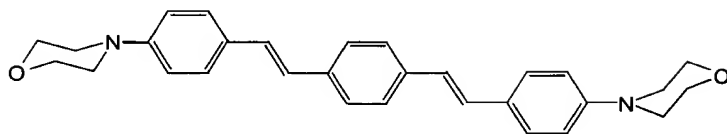




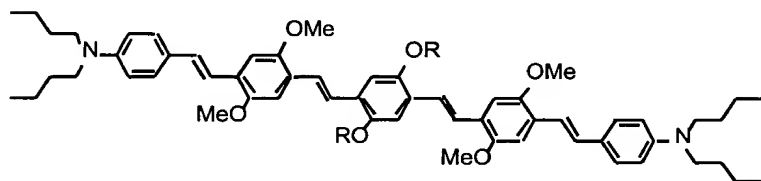
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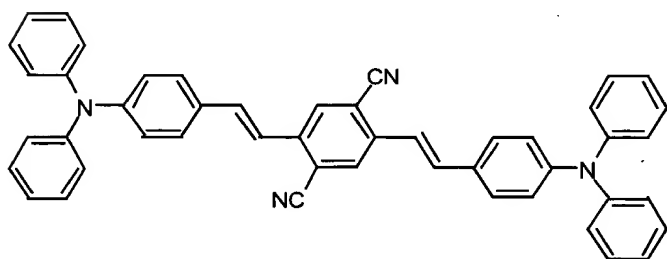
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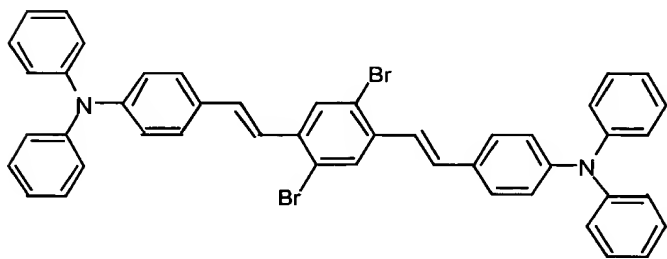
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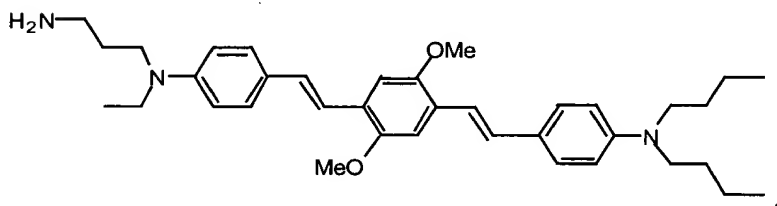
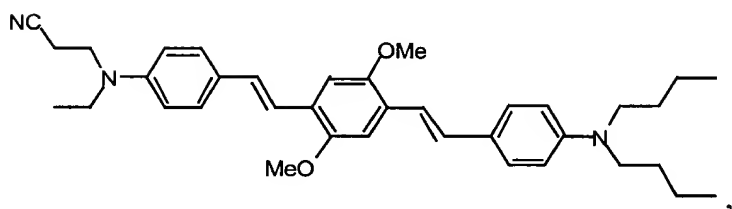
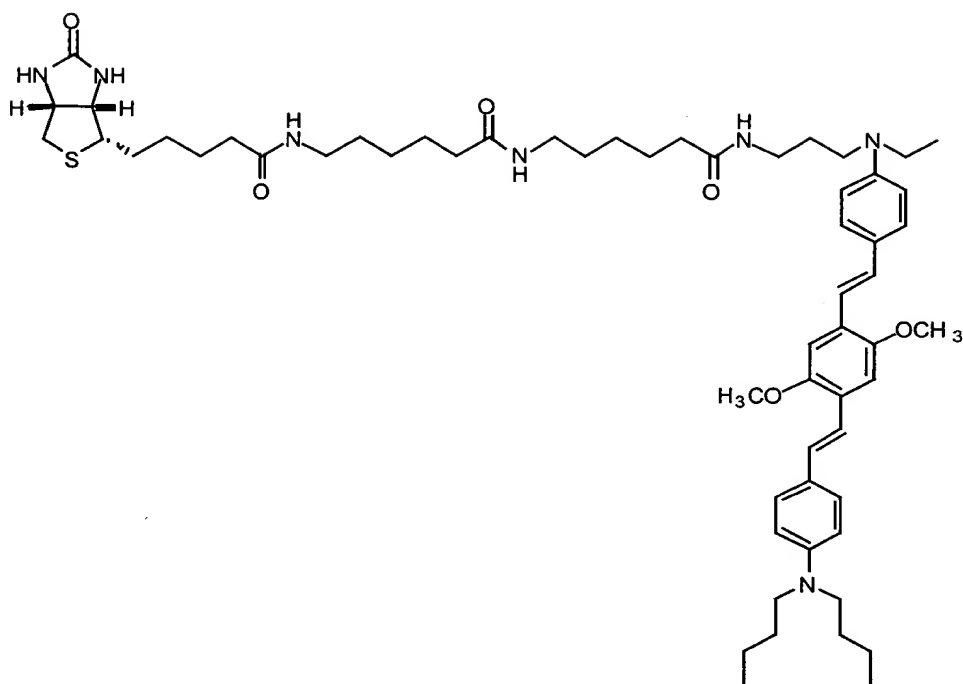
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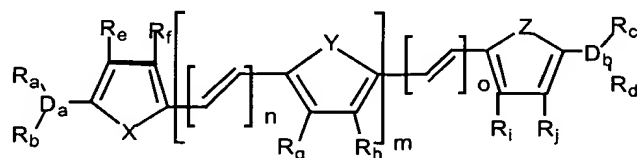
and mixtures thereof, where $R=(CH_2)_{11}CH_3$.

4. (Amended) A method for preparing a compound in an electronically excited state, comprising the steps of:

a) exposing a compound having the formula D_1-II-D_2 to radiation, wherein D_1 and D_2 are electron donor groups; and II comprises a bridge of π -conjugated bonds connecting D_1 and D_2 ; and

b) converting said compound to a multi-photon electronically excited state upon simultaneous absorption of at least two photons of said radiation by said compound, wherein the sum of the energies of all of said absorbed photons is greater than or equal to the

transition energy from a ground state of said compound to said multi-photon excited state and wherein the energy of each absorbed photon is less than the transition energy between said ground state and the lowest single-photon excited state of said compound and is less than the transition energy between said multi-photon excited state and said ground state [A method according to claim 2], wherein said compound is further defined by a formula



where D_a is selected from the group consisting of N, O, S and P;

where D_b is selected from the group consisting of N, O, S and P;

m, n, o are integers such that $0 \leq m \leq 10$, $0 \leq n \leq 10$, $0 \leq o \leq 10$; and

where:

X, Y, Z are independently selected from the group consisting of: $CR_k=CR_l$; O; S; and N- R_m ;

R_a, R_b, R_c, R_d are independently selected from the group consisting of: H; a linear or branched alkyl group with up to 25 carbons; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta OR_{a1}$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta NR_{a2}R_{a3}$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta CONR_{a2}R_{a3}$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta CN$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta Cl$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta Br$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta I$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta$ -Phenyl; where $0 < \alpha < 10$ and $1 < \beta < 25$, a group of aromatic rings having up to 20 carbons in the aromatic ring framework; fused aromatic rings, vinyl; allyl; 4-styryl; acrolyl; methacroyl; acrylonitrile, isocyanate; isothiocyanate; epoxides; strained ring olefins; $(-CH_2)_\delta SiCl_3$; $(-CH_2)_\delta Si(OCH_2CH_3)_3$; and $(-CH_2)_\delta Si(OCH_3)_3$; where $\delta < 25$;

$R_{a1}, R_{a2},$ and R_{a3} are independently selected from the group consisting of: H; a linear or branched alkyl group with up to 25 carbons, a functional group derived from an amino acid; a polypeptide; adenine; guanine; tyrosine; cytosine; uracil; biotin; ferrocene, ruthenocene, cyanuric chloride and derivatives thereof, or methacryloyl chloride;

$R_e, R_f, R_g, R_h, R_i, R_j, R_k, R_l$ and R_m are independently selected from the group consisting of: H; a linear or branched alkyl group with up to 25 carbons; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta OR_{b1}$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta NR_{b2}R_{b3}$;

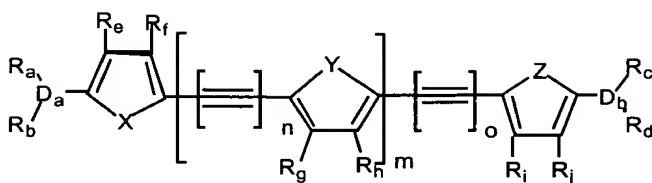
$-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{CONR}_{b2}\text{R}_{b3}$, where R_{b1} , R_{b2} , and R_{b3} are independently selected from the group consisting of a functional group derived from an amino acid; a polypeptide; adenine; guanine; tyrosine; cytosine; uracil; biotin; ferrocene, ruthenocene, cyanuric chloride and derivatives thereof or methacryloyl chloride; $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{CN}$;
 $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{Cl}$; $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{Br}$; $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{I}$;
 $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{-Phenyl}$, where $0 < \alpha < 10$ and $1 < \beta < 25$; a group of aromatic rings having up to 20 carbons in the aromatic framework; fused aromatic rings; CHO; CN; NO₂; Br; Cl; I; phenyl; an acceptor group containing more than two carbon atoms; a functional group derived from an amino acid and $\text{NR}_{e1}\text{R}_{e2}$; OR_{e3} ; where R_{e1} , R_{e2} , R_{e3} are defined as for R_n and R_o , where R_n and R_o are defined as any member of the group consisting of H; a linear or branched alkyl group with up to 25 carbons; $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{OR}_{g1}$;
 $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{NR}_{g2}\text{R}_{g3}$; $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{CONR}_{g2}\text{R}_{g3}$;
 $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{CN}$; $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{Cl}$; $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{Br}$;
 $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{I}$; $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{-Phenyl}$; aryl groups; fused aromatic rings; polymerizable functionalities;

R_{g1} , R_{g2} , and R_{g3} are independently selected from: H; a linear or branched alkyl group with up to 25 carbons; a functional group derived from an amino acid: a polypeptide; adenine; guanine; tyrosine; cytosine; uracil; biotin; ferrocene, ruthenocene, cyanuric chloride and derivatives thereof or methacryloyl chloride.

5. (Amended) A method for preparing a compound in an electronically excited state, comprising the steps of:

a) exposing a compound having the formula $\text{D}_1\text{-}\Pi\text{-}\text{D}_2$ to radiation, wherein D_1 and D_2 are electron donor groups; and Π comprises a bridge of π -conjugated bonds connecting D_1 and D_2 ; and

b) converting said compound to a multi-photon electronically excited state upon simultaneous absorption of at least two photons of said radiation by said compound, wherein the sum of the energies of all of said absorbed photons is greater than or equal to the transition energy from a ground state of said compound to said multi-photon excited state and wherein the energy of each absorbed photon is less than the transition energy between said ground state and the lowest single-photon excited state of said compound and is less than the transition energy between said multi-photon excited state and said ground state [A method according to claim 2], wherein said compound is further defined by a formula



where D_a is selected from the group consisting of N, O, S and P;

where D_b is selected from the group consisting of N, O, S and P;

m, n, o are integers such that $0 \leq m \leq 10$, $0 \leq n \leq 10$, $0 \leq o \leq 10$; and

where:

X, Y, Z are independently selected from the group consisting of: $CR_k=CR_l$; O; S; and $N-R_m$;

R_a, R_b, R_c, R_d are independently selected from the group consisting of: H; a linear or branched alkyl group with up to 25 carbons; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta OR_{a1}$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta NR_{a2}R_{a3}$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta CONR_{a2}R_{a3}$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta CN$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta Cl$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta Br$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta I$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta$ -Phenyl; where $0 < \alpha < 10$ and $1 < \beta < 25$, a group of aromatic rings having up to 20 carbons in the aromatic ring framework; fused aromatic rings, vinyl; allyl; 4-styryl; acryloyl; methacryloyl; acrylonitrile, isocyanate; isothiocyanate; epoxides; strained ring olefins; $(-CH_2)_\delta SiCl_3$; $(-CH_2)_\delta Si(OCH_2CH_3)_3$; and $(-CH_2)_\delta Si(OCH_3)_3$; where $\delta < 25$;

R_{a1}, R_{a2} , and R_{a3} are independently selected from the group consisting of: H; a linear or branched alkyl group with up to 25 carbons, a functional group derived from an amino acid; a polypeptide; adenine; guanine; tyrosine; cytosine; uracil; biotin; ferrocene, ruthenocene, cyanuric chloride and derivatives thereof, or methacryloyl chloride;

$R_e, R_f, R_g, R_h, R_i, R_j, R_k, R_l$ and R_m are independently selected from the group consisting of: H; a linear or branched alkyl group with up to 25 carbons; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta OR_{b1}$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta NR_{b2}R_{b3}$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta CONR_{b2}R_{b3}$, where R_{b1}, R_{b2} , and R_{b3} are independently selected from a functional group derived from an amino acid, a polypeptide; adenine; guanine; tyrosine; cytosine; uracil; biotin; ferrocene, ruthenocene, cyanuric chloride and derivatives thereof or methacryloyl chloride; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta CN$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta Cl$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta Br$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta I$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta$ -Phenyl, where $0 < \alpha < 10$ and $1 < \beta < 25$; a group of aromatic rings having up to 20 carbons in the aromatic

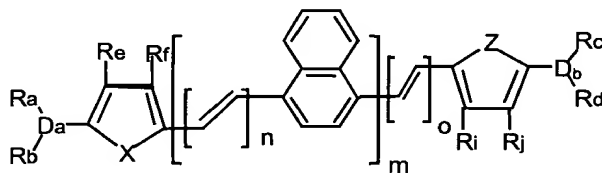
framework; fused aromatic rings; CHO; CN; NO₂; Br; Cl; I; phenyl; an acceptor group containing more than two carbon atoms; a functional group derived from an amino acid and NRe₁Re₂; ORe₃; where Re₁, Re₂, Re₃ are defined as for R_n and R_o, where R_n and R_o are defined as any member of the group consisting of H; a linear or branched alkyl group with up to 25 carbons; -(CH₂CH₂O)_α-(CH₂)_βOR_{g1}; -(CH₂CH₂O)_α-(CH₂)_βNR_{g2}R_{g3}; -(CH₂CH₂O)_α-(CH₂)_βCONR_{g2}R_{g3}; -(CH₂CH₂O)_α-(CH₂)_βCN; -(CH₂CH₂O)_α-(CH₂)_βCl; -(CH₂CH₂O)_α-(CH₂)_βBr; -(CH₂CH₂O)_α-(CH₂)_βI; -(CH₂CH₂O)_α-(CH₂)_β-Phenyl; aryl groups; fused aromatic rings; polymerizable functionalities;

R_{g1}, R_{g2}, and R_{g3} are independently selected from: H; a linear or branched alkyl group with up to 25 carbons; a functional group derived from an amino acid: a polypeptide; adenine; guanine; tyrosine; cytosine; uracil; biotin; ferrocene, ruthenocene, cyanuric chloride and derivatives thereof or methacryloyl chloride.

6. (Amended) A method for preparing a compound in an electronically excited state, comprising the steps of:

a) exposing a compound having the formula D₁-Π-D₂ to radiation, wherein D₁ and D₂ are electron donor groups; and Π comprises a bridge of π-conjugated bonds connecting D₁ and D₂; and

b) converting said compound to a multi-photon electronically excited state upon simultaneous absorption of at least two photons of said radiation by said compound, wherein the sum of the energies of all of said absorbed photons is greater than or equal to the transition energy from a ground state of said compound to said multi-photon excited state and wherein the energy of each absorbed photon is less than the transition energy between said ground state and the lowest single-photon excited state of said compound and is less than the transition energy between said multi-photon excited state and said ground state [A method according to claim 2], wherein said compound is further defined by a formula



where D_a is selected from the group consisting of N, O, S and P;

where D_b is selected from the group consisting of N, O, S and P;

m, n, o are integers such that $0 \leq m \leq 10$, $0 \leq n \leq 10$, $0 \leq o \leq 10$; and
where:

X, Y, Z are independently selected from the group consisting of: $CR_k=CR_l$; O; S;
and N- R_m ;

R_a , R_b , R_c , R_d are independently selected from the group consisting of: H; a linear
or branched alkyl group with up to 25 carbons; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta OR_{a1}$;
 $-(CH_2CH_2O)_\alpha-(CH_2)_\beta NR_{a2}R_{a3}$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta CONR_{a2}R_{a3}$;
 $-(CH_2CH_2O)_\alpha-(CH_2)_\beta CN$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta Cl$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta Br$;
 $-(CH_2CH_2O)_\alpha-(CH_2)_\beta I$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta$ -Phenyl; where $0 < \alpha < 10$ and $1 < \beta < 25$, a
group of aromatic rings having up to 20 carbons in the aromatic ring framework; fused
aromatic rings, vinyl; allyl; 4-styryl; acrolyl; methacroyl; acrylonitrile, isocyanate;
isothiocyanate; epoxides; strained ring olefins; $(-CH_2)_\delta SiCl_3$; $(-CH_2)_\delta Si(OCH_2CH_3)_3$; and
 $(-CH_2)_\delta Si(OCH_3)_3$; where $\delta < 25$;

R_{a1} , R_{a2} , and R_{a3} are independently selected from the group consisting of: H; a
linear or branched alkyl group with up to 25 carbons, a functional group derived from an
amino acid; a polypeptide; adenine; guanine; tyrosine; cytosine; uracil; biotin; ferrocene,
ruthenocene, cyanuric chloride and derivatives thereof, or methacryloyl chloride;

R_e , R_f , R_i , R_j , R_k , R_l and R_m are independently selected from the group consisting
of: H; a linear or branched alkyl group with up to 25 carbons; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta OR_{b1}$;
 $-(CH_2CH_2O)_\alpha-(CH_2)_\beta NR_{b2}R_{b3}$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta CONR_{b2}R_{b3}$, where R_{b1} , R_{b2} , and
 R_{b3} are independently selected from a functional group derived from an amino acid; a
polypeptide; adenine; guanine; tyrosine; cytosine; uracil; biotin; ferrocene, ruthenocene,
cyanuric chloride and derivatives thereof or methacryloyl chloride;
 $-(CH_2CH_2O)_\alpha-(CH_2)_\beta CN$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta Cl$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta Br$;
 $-(CH_2CH_2O)_\alpha-(CH_2)_\beta I$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta$ -Phenyl, where $0 < \forall < 10$ and $1 < \exists < 25$; a
group of aromatic rings having up to 20 carbons in the aromatic framework; fused aromatic
rings; CHO; CN; NO₂; Br; Cl; I; phenyl; an acceptor group containing more than two carbon
atoms; a functional group derived from an amino acid and $NR_{e1}R_{e2}$; OR_{e3} ; where R_{e1} , R_{e2} ,
 R_{e3} are defined as for R_n and R_o , where R_n and R_o are defined as any member of the group
consisting of H; a linear or branched alkyl group with up to 25 carbons;
 $-(CH_2CH_2O)_\alpha-(CH_2)_\beta OR_{g1}$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta NR_{g2}R_{g3}$;

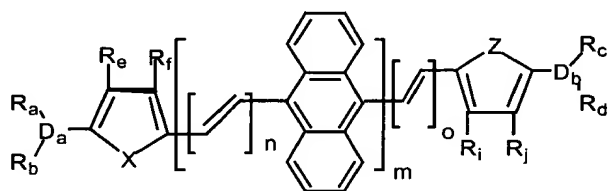
$-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{CONR}_{g2}\text{R}_{g3}$; $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{CN}$; $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{Cl}$;
 $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{Br}$; $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{I}$; $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{-Phenyl}$; aryl
 groups; fused aromatic rings; polymerizable functionalities;

R_{g1} , R_{g2} , and R_{g3} are independently selected from: H; a linear or branched alkyl group with up to 25 carbons; a functional group derived from an amino acid: a polypeptide; adenine; guanine; tyrosine; cytosine; uracil; biotin; ferrocene, ruthenocene, cyanuric chloride and derivatives thereof or methacryloyl chloride.

7. (Amended) A method for preparing a compound in an electronically excited state, comprising the steps of:

a) exposing a compound having the formula $\text{D}_1\text{-}\Pi\text{-D}_2$ to radiation, wherein D_1 and D_2 are electron donor groups; and Π comprises a bridge of π -conjugated bonds connecting D_1 and D_2 ; and

b) converting said compound to a multi-photon electronically excited state upon simultaneous absorption of at least two photons of said radiation by said compound, wherein the sum of the energies of all of said absorbed photons is greater than or equal to the transition energy from a ground state of said compound to said multi-photon excited state and wherein the energy of each absorbed photon is less than the transition energy between said ground state and the lowest single-photon excited state of said compound and is less than the transition energy between said multi-photon excited state and said ground state [A method according to claim 2], wherein said compound is further defined by a formula



where D_a is selected from the group consisting of N, O, S and P;

where D_b is selected from the group consisting of N, O, S and P;

m , n , o are integers such that $0 \leq m \leq 10$, $0 \leq n \leq 10$, $0 \leq o \leq 10$; and

where:

X, Y, Z are independently selected from the group consisting of: $\text{CR}_k=\text{CR}_l$; O; S; and N- R_m ;

R_a , R_b , R_c , R_d are independently selected from the group consisting of: H; a linear or branched alkyl group with up to 25 carbons; $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{OR}_{a1}$;

$-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{NR}_{a2}\text{R}_{a3}$; $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{CONR}_{a2}\text{R}_{a3}$;
 $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{CN}$; $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{Cl}$; $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{Br}$;
 $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{I}$; $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{-Phenyl}$; where $0 < \alpha < 10$ and $1 < \beta < 25$, a
 group of aromatic rings having up to 20 carbons in the aromatic ring framework; fused
 aromatic rings, vinyl; allyl; 4-styryl; acroyl; methacroyl; acrylonitrile, isocyanate;
 isothiocyanate; epoxides; strained ring olefins; $(-\text{CH}_2)_\delta\text{SiCl}_3$; $(-\text{CH}_2)_\delta\text{Si}(\text{OCH}_2\text{CH}_3)_3$; and
 $(-\text{CH}_2)_\delta\text{Si}(\text{OCH}_3)_3$; where $\delta < 25$;

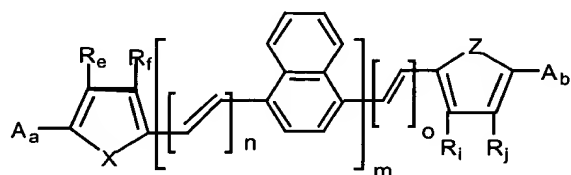
R_{a1} , R_{a2} , and R_{a3} are independently selected from the group consisting of: H; a
 linear or branched alkyl group with up to 25 carbons, a functional group derived from an
 amino acid; a polypeptide; adenine; guanine; tyrosine; cytosine; uracil; biotin; ferrocene,
 ruthenocene, cyanuric chloride and derivatives thereof, or methacryloyl chloride;

R_e , R_f , R_i , R_j , R_k , R_l and R_m are independently selected from the group consisting
 of: H; a linear or branched alkyl group with up to 25 carbons; $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{OR}_{b1}$;
 $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{NR}_{b2}\text{R}_{b3}$; $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{CONR}_{b2}\text{R}_{b3}$, where R_{b1} , R_{b2} , and
 R_{b3} are independently selected from the group consisting of a functional group derived from
 an amino acid; a polypeptide; adenine; guanine; tyrosine; cytosine; uracil; biotin; ferrocene,
 ruthenocene, cyanuric chloride and derivatives thereof or methacryloyl chloride;
 $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{CN}$; $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{Cl}$; $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{Br}$;
 $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{I}$; $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{-Phenyl}$, where $0 < \alpha < 10$ and $1 < \beta < 25$; a
 group of aromatic rings having up to 20 carbons in the aromatic framework; fused aromatic
 rings; CHO; CN; NO₂; Br; Cl; I; phenyl; an acceptor group containing more than two carbon
 atoms; a functional group derived from an amino acid and $\text{NR}_{e1}\text{R}_{e2}$; OR_{e3} ; where R_{e1} , R_{e2} ,
 R_{e3} are defined as for R_n and R_o , where R_n and R_o are defined as any member of the group
 consisting of H; a linear or branched alkyl group with up to 25 carbons;
 $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{OR}_{g1}$; $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{NR}_{g2}\text{R}_{g3}$;
 $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{CONR}_{g2}\text{R}_{g3}$; $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{CN}$; $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{Cl}$;
 $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{Br}$; $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{I}$; $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{-Phenyl}$; aryl
 groups; fused aromatic ring; polymerizable functionalities;
 R_{g1} , R_{g2} , and R_{g3} are independently selected from: H; a linear or branched alkyl group with
 up to 25 carbons; a functional group derived from an amino acid; a polypeptide; adenine;
 guanine; tyrosine; cytosine; uracil; biotin; ferrocene, ruthenocene, cyanuric chloride and
 derivatives thereof or methacryloyl chloride.

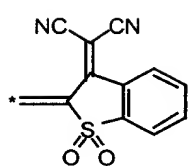
13. (Amended) A method for preparing a compound in an electronically excited state, comprising the steps of:

a) exposing a compound having the formula A_1 - Π - A_2 to radiation, wherein A_1 and A_2 are electron acceptors; and Π comprises a bridge of π -conjugated bonds connecting A_1 and A_2 ; and

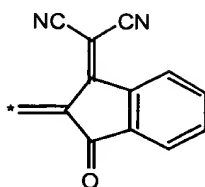
b) converting said compound to a multi-photon electronically excited state upon simultaneous absorption of at least two photons of said radiation by said compound, wherein the sum of the energies of all of said absorbed photons is greater than or equal to the transition energy from a ground state of said compound to said multi-photon excited state and wherein the energy of each absorbed photon is less than the transition energy between said ground state and the lowest single-photon excited state of said compound and is less than the transition energy between said multi-photon excited state and said ground state [A method according to claim 9], wherein said compound is further defined by a formula



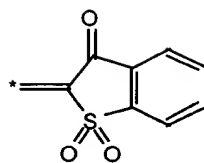
where A_a and A_b can be independently selected from: CHO; CN; NO₂, and



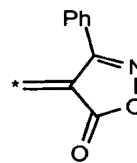
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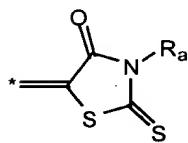
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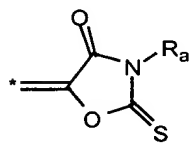
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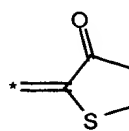
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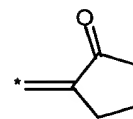
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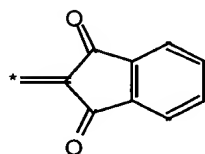
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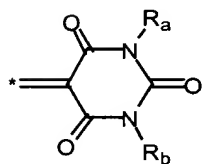
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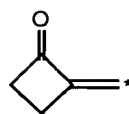
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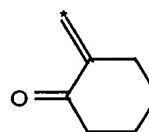
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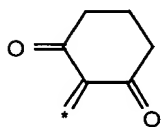
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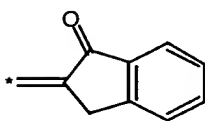
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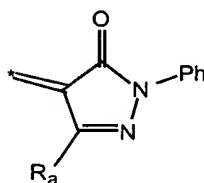
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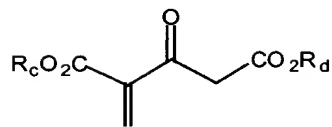
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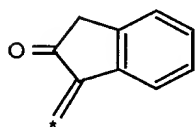
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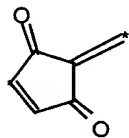
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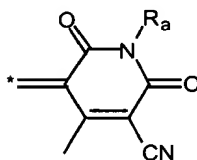
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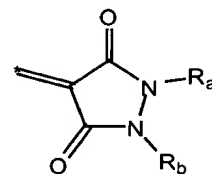
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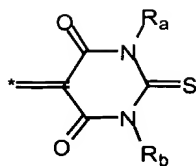
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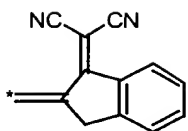
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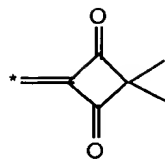
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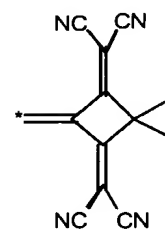
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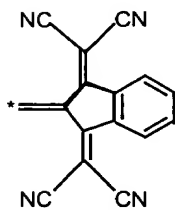
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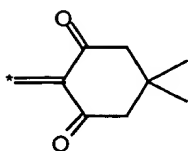
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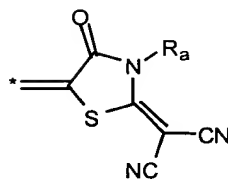
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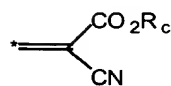
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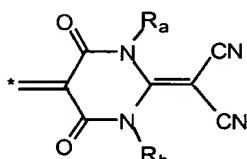
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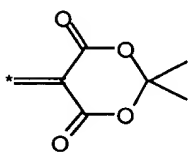
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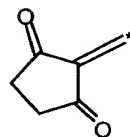
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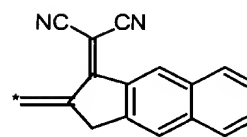
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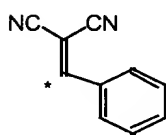
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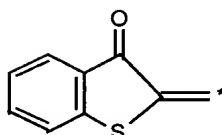
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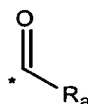
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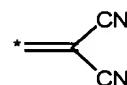
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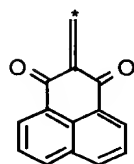
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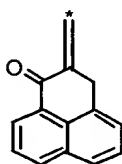
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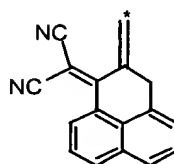
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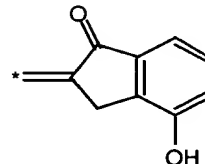
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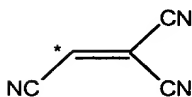
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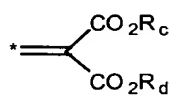
A39



A40



A41



A42

in addition A_a and A_b can be independently selected from Br, Cl, and I; and where $0 \leq m \leq 10$, $0 \leq n \leq 10$, $0 \leq o \leq 10$ [.]; and where:

X, Y, Z are independently selected from the group consisting of: $CR_k=CR_l$; O; S; and $N-R_m$;

R_a , R_b , R_c , R_d are independently selected from the group consisting of: H; a linear or branched alkyl group with up to 25 carbons; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta OR_{a1}$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta NR_{a2}R_{a3}$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta CONR_{a2}R_{a3}$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta CN$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta Cl$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta Br$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta I$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta$ -Phenyl; where $0 < \alpha < 10$ and $1 < \beta < 25$, a group of aromatic rings having up to 20 carbons in the aromatic ring framework; fused aromatic rings, vinyl; allyl; 4-styryl; acrolyl; methacroyl; acrylonitrile, isocyanate; isothiocyanate; epoxides; strained ring olefins; $-(CH_2)_\delta SiCl_3$; $-(CH_2)_\delta Si(OCH_2CH_3)_3$; and $-(CH_2)_\delta Si(OCH_3)_3$; where $\delta < 25$;

R_{a1} , R_{a2} , and R_{a3} are independently selected from the group consisting of: H; a linear or branched alkyl group with up to 25 carbons, a functional group derived from an amino acid; a polypeptide; adenine; guanine; tyrosine; cytosine; uracil; biotin; ferrocene, ruthenocene, cyanuric chloride and derivatives thereof, or methacryloyl chloride;

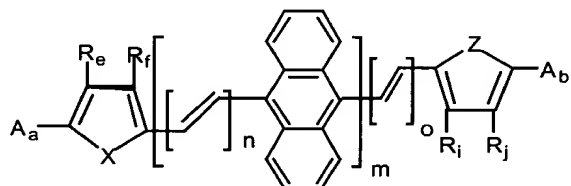
R_e , R_f , R_i , R_j , R_k , R_l and R_m are independently selected from the group consisting of: H; a linear or branched alkyl group with up to 25 carbons; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta OR_{b1}$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta NR_{b2}R_{b3}$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta CONR_{b2}R_{b3}$, where R_{b1} , R_{b2} , and R_{b3} are independently selected from the group consisting of a functional group derived from an amino acid; a polypeptide; adenine; guanine; tyrosine; cytosine; uracil; biotin; ferrocene, ruthenocene, cyanuric chloride and derivatives thereof or methacryloyl chloride; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta CN$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta Cl$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta Br$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta I$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta$ -Phenyl, where $0 < \forall < 10$ and $1 < \exists < 25$; a group of aromatic rings having up to 20 carbons in the aromatic framework; fused aromatic rings; CHO; CN; NO₂; Br; Cl; I; phenyl; an acceptor group containing more than two carbon atoms; a functional group derived from an amino acid and $NR_{e1}R_{e2}$; OR_{e3} ; where R_{e1} , R_{e2} , R_{e3} are defined as for R_n and R_o , where R_n and R_o are defined as any member of the group consisting of H; a linear or branched alkyl group with up to 25 carbons; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta OR_{g1}$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta NR_{g2}R_{g3}$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta CONR_{g2}R_{g3}$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta CN$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta Cl$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta Br$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta I$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta$ -Phenyl; aryl groups; fused aromatic rings; polymerizable functionalities;

R_{g1} , R_{g2} , and R_{g3} are independently selected from: H; a linear or branched alkyl group with up to 25 carbons; a functional group derived from an amino acid; or a polypeptide; adenine; guanine; tyrosine; cytosine; uracil; biotin; ferrocene, ruthenocene, cyanuric chloride and derivatives thereof or methacryloyl chloride.

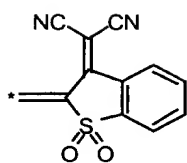
14. (Amended) A method for preparing a compound in an electronically excited state, comprising the steps of:

a) exposing a compound having the formula A_1 - Π - A_2 to radiation, wherein A_1 and A_2 are electron acceptors; and Π comprises a bridge of π -conjugated bonds connecting A_1 and A_2 ; and

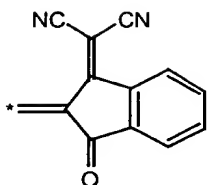
b) converting said compound to a multi-photon electronically excited state upon simultaneous absorption of at least two photons of said radiation by said compound, wherein the sum of the energies of all of said absorbed photons is greater than or equal to the transition energy from a ground state of said compound to said multi-photon excited state and wherein the energy of each absorbed photon is less than the transition energy between said ground state and the lowest single-photon excited state of said compound and is less than the transition energy between said multi-photon excited state and said ground state [A method according to claim 9], wherein said compound is further defined by a formula



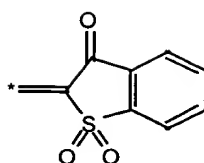
where A_a and A_b can be independently selected from: CHO; CN; NO_2 , and



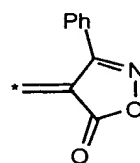
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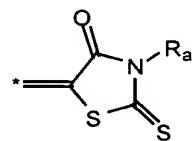
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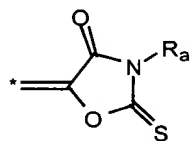
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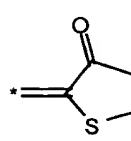
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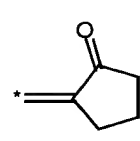
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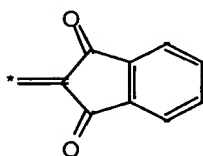
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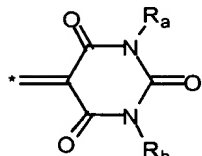
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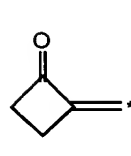
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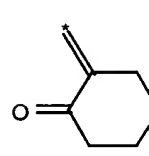
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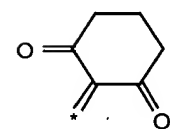
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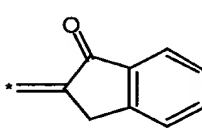
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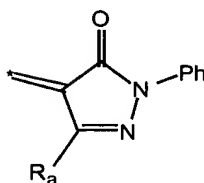
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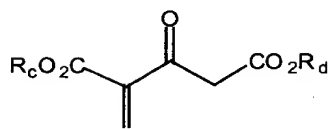
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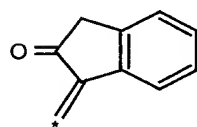
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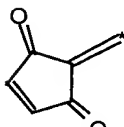
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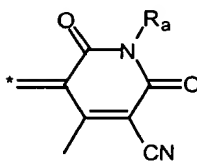
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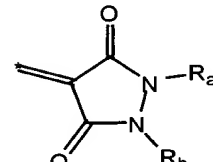
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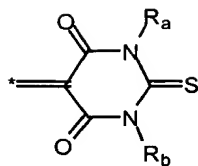
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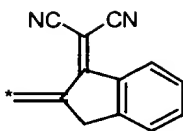
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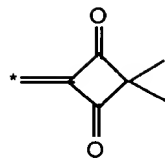
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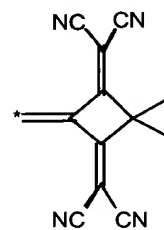
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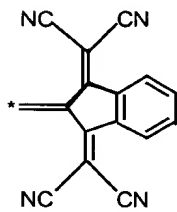
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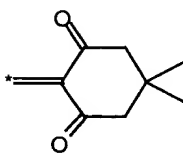
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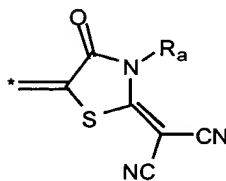
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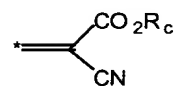
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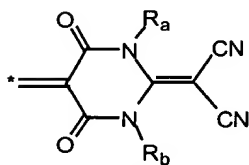
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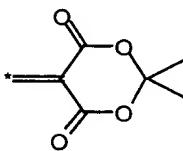
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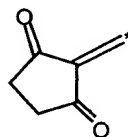
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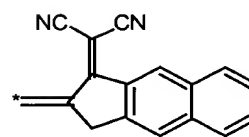
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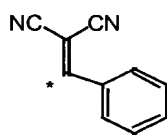
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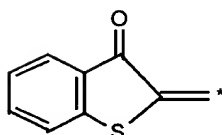
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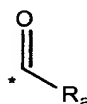
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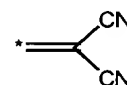
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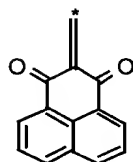
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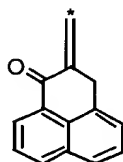
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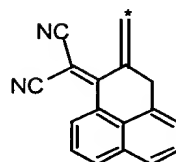
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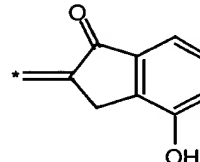
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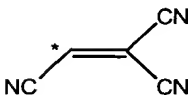
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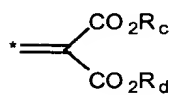
A39



A40



A41



A42

in addition A_a and A_b can be independently selected from Br, Cl, and I; and where $0 \leq m \leq 10$, $0 \leq n \leq 10$, $0 \leq o \leq 10$ [.]; and where:

X, Y, Z are independently selected from the group consisting of: $CR_k=CR_l$; O; S; and N- R_m ;

R_a, R_b, R_c, R_d are independently selected from the group consisting of: H; a linear or branched alkyl group with up to 25 carbons; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta OR_{a1}$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta NR_{a2}R_{a3}$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta CONR_{a2}R_{a3}$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta CN$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta Cl$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta Br$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta I$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta$ -Phenyl; where $0 < \alpha < 10$ and $1 < \beta < 25$, a group of aromatic rings having up to 20 carbons in the aromatic ring framework; fused aromatic rings, vinyl; allyl; 4-styryl; acrolyl; methacroyl; acrylonitrile, isocyanate; isothiocyanate; epoxides; strained ring olefins; $(-CH_2)_\delta SiCl_3$; $(-CH_2)_\delta Si(OCH_2CH_3)_3$; and $(-CH_2)_\delta Si(OCH_3)_3$; where $\delta < 25$;

R_{a1}, R_{a2} , and R_{a3} are independently selected from the group consisting of: H; a linear or branched alkyl group with up to 25 carbons, a functional group derived from an amino acid; a polypeptide; adenine; guanine; tyrosine; cytosine; uracil; biotin; ferrocene, ruthenocene, cyanuric chloride and derivatives thereof, or methacryloyl chloride;

$R_e, R_f, R_i, R_j, R_k, R_l$ and R_m are independently selected from the group consisting of: H; a linear or branched alkyl group with up to 25 carbons; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta OR_{b1}$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta NR_{b2}R_{b3}$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta CONR_{b2}R_{b3}$, where R_{b1}, R_{b2} , and R_{b3} are independently selected from the group consisting of a functional group derived from an amino acid; a polypeptide; adenine; guanine; tyrosine; cytosine; uracil; biotin; ferrocene, ruthenocene, cyanuric chloride and derivatives thereof or methacryloyl chloride; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta CN$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta Cl$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta Br$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta I$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta$ -Phenyl, where $0 < \forall < 10$ and $1 < \exists < 25$; a group of aromatic rings having up to 20 carbons in the aromatic framework; fused aromatic rings; CHO; CN; NO₂; Br; Cl; I; phenyl; an acceptor group containing more than two carbon atoms; a functional group derived from an amino acid and $NR_{e1}R_{e2}$; OR_{e3} ; where R_{e1}, R_{e2}, R_{e3} are defined as for R_n and R_o , where R_n and R_o are defined as any member of the group consisting of H; a linear or branched alkyl group with up to 25 carbons; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta OR_{g1}$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta NR_{g2}R_{g3}$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta CONR_{g2}R_{g3}$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta CN$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta Cl$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta Br$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta I$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta$ -Phenyl; aryl groups; fused aromatic rings; polymerizable functionalities;

R_{g1} , R_{g2} , and R_{g3} are independently selected from: H; a linear or branched alkyl group with up to 25 carbons; a functional group derived from an amino acid; or a polypeptide; adenine; guanine; tyrosine; cytosine; uracil; biotin; ferrocene, ruthenocene, cyanuric chloride and derivatives thereof or methacryloyl chloride.



Appendix C

MARKED UP VERSION OF AMENDED PARAGRAPHS

Note: Amended paragraphs are preceded by the location of their first line, indicated in **bold type**. Additions to the paragraphs are underlined. Deletions appear in [brackets].

Page 1, line 16:

This application is a divisional application of U.S. Application No. 08/965,945 filed November 7, 1997, now U.S. Patent No. 6,267,913 and claims the benefit of U.S. Provisional Application No. 60/029,443 entitled "Two-Photon Optical Limiting Materials," U.S. Provisional Application No. 60/029,437, entitled "Two-Photon Absorbing Polymerization Initiators" and U.S. Provisional Application No. 60/030,141, entitled "Two-Photon Absorbing Optical Materials", all filed November 12, 1996, the disclosure of each of which is incorporated herein by reference.

Page 9, line 13:

By the term "chromophore", it is meant a molecule, a molecular fragment, or aggregate of molecules that can absorb electromagnetic radiation.

Page 9, line 25:

By the phrase "optical limiting", it is meant a process wherein the optical transmission of a body decreases with increasing incident optical intensity or fluence such that the intensity [of]or fluence transmitted by the body does not exceed a prescribed level for a prescribed range of incident optical intensity or fluence.

Page 10, line 19:

The invention generally provides compositions of matter that have large two-photon or higher-order absorptivities and in some cases large fluorescence quantum yields, leading to efficient two-photon or higher-order excited fluorescence. By two-photon or higher-order absorption, we refer to the initial simultaneous absorption of two or more photons (also referred to as multi-photon absorption), without the actual population of an excited state by the absorption of a single photon. [That is, subsequent]Subsequent to the initial absorption of two or more photons by the molecule, it is possible for the molecule to absorb additional photons from the state populated by the initial absorption event or from a state to which the

molecule relaxes. If sufficient total energy is absorbed by the molecule, it is possible to photoionize the molecule, thereby creating a radical cation of the molecule and an unbound electron. Conversion of a molecule to an excited state by two-photon or higher-order absorption according to the invention enables many applications which can be induced from such an excited state, as described below.

Page 11, line 3:

For example, we have found that stilbene, diphenylpolyene, phenylene vinylene oligomers, and related molecules having two or more electron donor groups, such as amino groups or alkoxy groups, connected to aromatic or heteroaromatic end groups as part of a π (pi)-electron bridge (the combination referred to herein as “D- π -D” molecules) exhibit unexpectedly and unusually high two-photon or higher-order absorptivities in comparison to unsubstituted dyes such as stilbene, diphenylpolyenes, phenylene vinylene oligomers and related molecules, respectively. In addition, the strength and position of the two-photon or higher-order absorption of these molecules can be tuned and further enhanced by appropriate substitution of the π -electron bridge with at least one electron accepting group, such as cyano, (referred to herein as “D-A-D” molecules). We have further discovered that molecules having two or more electron acceptor groups such as formyl or dicyanomethylidene groups, connected to aromatic or heteroaromatic end groups as part of a π (pi)-electron bridge (referred to herein as “A- π -A” molecules) exhibit unexpectedly and unusually high two-photon or higher-order absorptivities in comparison to dyes such as stilbene, diphenylpolyenes, phenylene vinylene oligomers and related molecules. The strength and position of the two-photon or higher-order absorption of these molecules can be tuned and further enhanced by appropriate substitution of the π -electron bridge with electron donating groups, such as methoxy, to form “A-D-A” molecules.

Page 11, line 24:

As a feature of the invention, we have discovered that the aromaticity of the π -bridge in the electronic ground state is important in determining the energetic position of the electronic state which is responsible for multi-photon absorption relative to the state responsible for one-photon absorption. In particular, having donors attached to aromatic groups or heteroaromatic groups, i.e., D- π -D and D-A-D structures, is conducive to [placing]the occurrence of the highly absorbing two-photon state energetically above that of

the strongly fluorescent one-photon state, which is essential for the molecule to exhibit a high multi-photon fluorescence excitation efficiency. In D-A-D molecules, upon absorption of photons, charge is redistributed from the electron donor groups toward the π -electron bridge, thereby enhancing the molecules' photon absorption characteristics.

Page 12, line 6:

In addition, having acceptors attached to aromatic groups or heteroaromatic groups, i.e., A- π -A and A-D-A structures, is conducive to [placing] the occurrence of the highly absorbing two-photon state energetically above that of the strongly fluorescent one-photon state. In A-D-A molecules, upon absorption of photons, charge is transferred from the π -electron bridge toward the electron accepting groups, thereby enhancing the molecules' photon absorption characteristics.

Page 12, line 12:

Accordingly, the combination of the intrinsically high fluorescence quantum efficiency for these molecules and the rapid relaxation of the two-photon excited molecule from the two-photon state to the lower lying fluorescent one-photon state makes the molecules according to the invention highly efficient multi-photon excitable fluorescent emitters.

Page 14, line 21:

These new two-photon or multi-photon absorbers may be used for two-photon or multi-photon two-dimensional or three-dimensional optical lithography and three dimensional optical memory.

Page 15, line 19:

Upon multi-photon excitation, these molecules become very strong excited state reducing agents and can therefore be used as multi-photon excited electron donors. Such materials can transfer electrons to electron acceptors (which may be covalently attached to the multi-photon absorbing chromophore or could be in a region of space near the multi-photon absorbing chromophore, as would be the case in a concentrated fluid solution or solid solution) to create a radical cation, in the case of [a] the multi-photon absorbing dye and a radical anion, in the case of the acceptor.

Page 15, line 27:

Another feature of some molecules in one embodiment of the invention is that molecules "end capped" with electron accepting substituents, i.e., A- π -A and A-D-A structures, have relatively high electron affinities in the ground-state form. Furthermore, upon multi-photon excitation, these molecules become very strong excited state oxidizing agents and can therefore be used as multi-photon excited electron acceptors. Such materials can accept electrons from electron donors (which may be covalently attached to the multi-photon absorbing chromophore or could be in a region of space near the multi-photon absorbing chromophore as would be the case in a concentrated solution or solid solution) to create a radical anion, in the case of [a]the multi-photon absorbing dye, and a radical cation, in the case of the donor.

Page 16, line 11:

In addition, these radical ion species generated upon multi-photon absorption can be chemical reactants. For example, these multi-photon absorbing materials can be used to initiate polymerization of monomers which can be polymerized either by radical or ionic mechanisms in the presence or absence of polymerization promoters, including chain transfer agents. Examples of monomers which can be polymerized in these ways include, but are not limited to, vinyl monomers including acrylonitriles, acrylates, diacrylates, triacrylates, polyacrylates, styrenes, trifunctional isocyanurates substituted with acrylates, vinyl- and allyl-containing monomers, divinylbenzene and polymers which are end-capped with or have these groups attached to the carbon backbone such as polyurethanes, polyesters and polyolefins. [In addition,] Additional examples of polymerizable monomers may be found in G. Odian, *Principles of Polymerization*, Second Edition, John Wiley and Son, New York, 1981, page 181, which is incorporated herein by reference.

Page 18, line 1:

Another feature of this invention is that it is possible to tune the lipophilicity, hydrophilicity and overall solubility of the multi-photon absorbing chromophores over a very wide range by the appropriate substitution of the donor groups. For example, 4,4'-bis-dimethylaminostilbene and 4,4'-bis-(diphenyl)aminostilbene are sparingly soluble in nonpolar

organic solvents whereas 4,4'-bis(-di-*n*-butylamino)stilbene and 4,4'-bis(di-(4-*n*-butyl-phenyl)aminostilbene are very soluble, i.e., are lipophilic. In contrast, the hydrochloric acid adduct of the bis-lysyl ester of 4-diethylamino 4'-diethanolaminostilbene and the bis-lysyl ester of 1-(4-dimethylaminophenyl)-4-(4'-diethanolaminophenyl) buta-1,3-diene shown below are hydrophilic. In each case, the molecules maintain their fluorescence in organic and aqueous solution.

Page 19, line 28:

An advantage which is gained by the efficient multi-photon absorption processes enabled by molecules according to the invention is that it is possible for the absorption to occur in a region of space whose cross sectional area is smaller than that of the diffraction-limited spot size of the exciting beam of light. This is because, for example, a two-photon process is dependent on I^2 , where I is the intensity of the exciting beam. The diffraction-limited spot size for a Gaussian beam is given by:

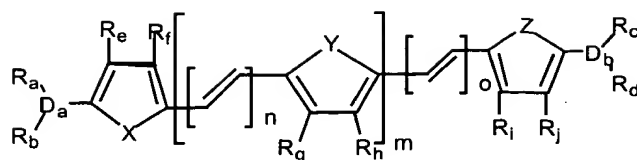
$$d_{1/e^2} = \frac{0.635\lambda}{NA}$$

where λ is the wavelength of the exciting beam of light and NA is the numerical aperture of the focusing lens, and d_{1/e^2} is the full width of the beam where the intensity is $1/e^2$ times the peak intensity. Therefore the theoretical spot size for a two-photon excitation is given by:

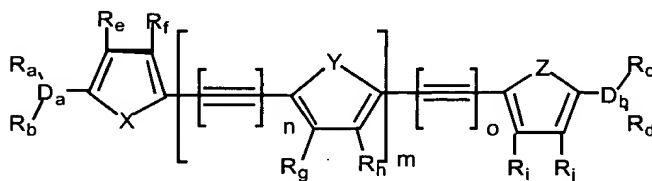
$$d_{\text{"2-photon"}} = \frac{d_{1/e^2}}{\sqrt{2}}$$

Page 21, line 6:

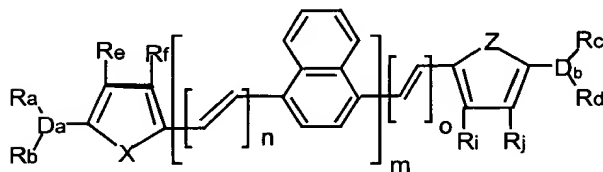
Class 1 Structures: Compounds where the end groups are electron donating groups



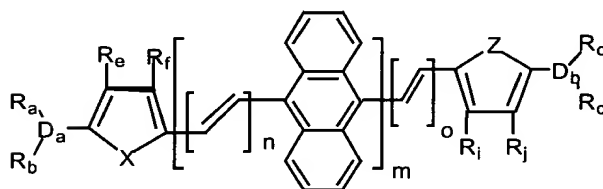
Class 1- I



Class 1- II



Class 1- III



Class 1- IV

where D_a is any electron donating group, that is any one of N, O, S and P;

where D_b is any electron donating group, that is any one of N, O, S and P;

m, n, o are integers such that $0 \leq m \leq 10$, $0 \leq n \leq 10$, $0 \leq o \leq 10$; and

where X, Y, Z are independently selected from: $CR_k=CR_l$; O; S; N- R_m

where $R_e, R_f, R_g, R_h, [R_i]R_l, R_j, R_k, R_l, R_m$ are defined in NOTE 3.

Page 22, line 8:

R_a, R_b, R_c, R_d are independently selected from: H; a linear or branched alkyl group with up to 25 carbons; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta CO_2 R_{a1}$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta OR_{a1}$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta NR_{a2} R_{a3}$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta CONR_{a2} R_{a3}$;

$-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{CN}$; $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{Cl}$; $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{Br}$;
 $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{I}$; $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{-Phenyl}$; various aryl groups (see NOTE 1);
 various fused aromatic rings (see NOTE 2); various polymerizable functionalities attached through a linkage which can be chosen from a linear or branched alkyl chain with up to 25 carbons, various aryl groups, $(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta-$, and $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{CO}_2$ (see NOTE 2A);

- R_{a1} , R_{a2} , and R_{a3} are independently selected from: H; a linear or branched alkyl group with up to 25 carbons;
- R_{a1} , R_{a2} , and R_{a3} can also be a functional group derived essentially from one of the 20 naturally occurring amino acids: alanine; valine; leucine; isoleucine; proline; tryptophan; phenylalanine; methionine; glycine; serine; threonine; tyrosine; cysteine; glutamine; asparagine; lysine; arginine; histidine; aspartic acid; and glutamic acid; a polypeptide; adenine; guanine; tyrosine; cytosine; uracil; biotin; ferrocene, ruthenocene, cyanuric chloride and derivatives thereof, or methacryloyl chloride. By “derived essentially”, it is meant that R_{a1} , R_{a2} , and R_{a3} may be an amino acid or a functional group that is closely related (e.g., a trivial modification of an amino acid).
- where α is 0-10,
- where β is 1- 25.

Page 24, line 18:

R_e , R_f , R_g , R_h , R_i , R_j , R_k , R_l , R_m are independently selected from: H; a linear or branched alkyl group with up to 25 carbons; $(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{-CO}_2R_{a1}$; -
 $(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{OR}_{b1}$;
 $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{NR}_{b2}R_{b3}$; $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{CONR}_{b2}R_{b3}$;
 $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{CN}$; $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{Cl}$; $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{Br}$;
 $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{I}$; $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{-Phenyl}$; various aryl groups (see NOTE 1);
 various fused aromatic rings (see NOTE 2); various polymerizable functionalities (see NOTE 2A); $\text{NR}_{e1}R_{e2}$; OR_{e3} ; CHO; CN; NO₂; Br; Cl; I; phenyl; an acceptor group containing more than two carbon atoms (see NOTE 3A), and where R_{e1} , R_{e2} , R_{e3} , are independently [as defined]defined as for R_n and R_o in NOTE 3B;

- R_{b1} , R_{b2} , and R_{b3} can also be a functional group derived essentially from one of the 20 naturally occurring amino acids: alanine; valine; leucine; isoleucine; proline; tryptophan; phenylalanine; methionine; glycine; serine; threonine; tyrosine; cysteine;

glutamine; asparagine; lysine; arginine; histidine; aspartic acid; and glutamic acid; or a polypeptide; adenine; guanine; tyrosine; cytosine; uracil; biotin; ferrocene; ruthenocene, cyanuric chloride and derivatives thereof or methacryloyl chloride. By “derived essentially from”, it is meant that R_{b1} , R_{b2} , and R_{b3} can be an amino acid or a functional group that is closely related to an amino acid (i.e., a trivial modification of an amino acid).

- where α is 0-10,
- where β is 1-25.

Page 27, line 5:

R_n , R_o are independently selected from: H; a linear or branched alkyl group with up to 25 carbons; $(CH_2CH_2O)_\alpha-(CH_2)_\beta-CO_2R_{\alpha1}$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta OR_{g1}$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta NR_{g2}R_{g3}$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta CONR_{g2}R_{g3}$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta CN$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta Cl$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta Br$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta I$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta$ -Phenyl; various aryl groups (see NOTE 1); various fused aromatic rings (see NOTE 2); various polymerizable functionalities (see NOTE 2A);

R_{g1} , R_{g2} , and R_{g3} are independently selected from: H; a linear or branched alkyl group with up to 25 carbons;

- R_{g1} , R_{g2} , and R_{g3} can also be a functional group derived essentially from one of the 20 naturally occurring amino acids: alanine; valine; leucine; isoleucine; proline; tryptophan; phenylalanine; methionine; glycine; serine; threonine; tyrosine; cysteine; glutamine; asparagine; lysine; arginine; histidine; aspartic acid; and glutamic acid; or a polypeptide; adenine; guanine; tyrosine; cytosine; uracil; biotin; ferrocene, ruthenocene, cyanuric chloride and derivatives thereof or methacryloyl chloride. By “derived essentially from” it is meant that R_{g1} , R_{g2} , and R_{g3} can be an amino acid or a functional group that is closely related to an amino acid (i.e., a trivial modification of an amino acid).
- where α is 0-10, and where β is 1- 25.

Page 31, line 7:

R_a , R_b , R_c , R_d are independently selected from: H; a linear or branched alkyl group with up to 25 carbons; $(CH_2CH_2O)_\alpha-(CH_2)_\beta-CO_2R_{\alpha1}$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta OR_{a1}$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta NR_{a2}R_{a3}$; $-(CH_2CH_2O)_\alpha-(CH_2)_\beta CONR_{a2}R_{a3}$;

$-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{CN}$; $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{Cl}$; $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{Br}$; $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{I}$; $-(\text{CH}_2\text{CH}_2\text{O})_\alpha-(\text{CH}_2)_\beta\text{-Phenyl}$; various aryl groups (see NOTE 1 as described above for Class 1 molecules); various fused aromatic rings (see NOTE 2 as described above for Class 1 molecules); various polymerizable functionalities (see NOTE 2A as described above for Class 1 molecules); $\text{NR}_{e1}\text{R}_{e2}$; OR_{e3} ; CN ; NO_2 ; Br ; Cl ; I ; phenyl; or an acceptor group containing more than two carbon atoms (see NOTE 3A as described above for Class 1 molecules).

Page 32, line 3:

The compositions of the invention are organic molecules having multi-photon absorption properties. The compositions themselves may exist as crystals, mesoscopic phases, polymers, glasses, liquids or gases. The compositions may be used alone or in combination with other crystals, mesoscopic phases, polymers, glasses liquids or gases.

Page 33, line 3:

Generally, the methods according to invention are carried out by converting a multi-photon absorbing molecule to an electronically excited state by absorption of at least two photons of radiation. The excited state then facilitates numerous applications. [In addition, the]The molecule may be irradiated with visible, ultraviolet or infrared radiation to effect the multiphoton absorption.

Page 54, line 3:

0.01 mol of the appropriate dialcohol, 0.03 mol of the amino acid and 0.025 mol of hydroxybenztriazole ($\text{HOBT}\cdot\text{H}_2\text{O}$) were dissolved in 60 mL of anhydrous N,N-dimethylformamide and stirred under nitrogen atmosphere. After cooling down to 0°C , 0.035 mol of dicyclohexylcarbodiimide were added at once. The solution was allowed to warm to ambient temperature and stirred for further 15 h. The precipitated urea was filtered off and the solution was diluted with 200 mL of diethyl ether. The organic phase was washed three times with 50 mL of water, dried over Na_2SO_4 and evaporated to dryness. To remove [of] the excess of amino acid, the residue was dissolved in diethyl ether and filtered over a short

column of Florisil. The organic solvent was evaporated again yielding the bisester as a solid. Yields were typically around 70-80%.

Page 72, line 7:

1,4-Bis(4-morpholino)styryl Benzene : prepared from the reaction of 4-morpholinobenzaldehyde (3.94 g, 0.02 mol) and 1,4-bis(methyltriphenylphosphonium chloride) (8.42g, 0.012 mol) and lithium/ethanol (0.296g, 0.043 mol/ 150 mL). Recrystallized from ethanol to yield a mixture of isomeric products, Yield 8%; Recrystallized from acetone to yield one pure isomer, Yield 11.8%; m.p. = 155°C; IR (KBr) ν cm^{-1} : 1604, 1519, 1355, 1179, 962, 822; UV-Vis (acetone) λ_{max} , nm: 400; ^1H NMR (CDCl_3 , δ): 3.66 (t, J = 6.9 Hz, 8H), 3.76 (t, J = 6.9 Hz, 8H), 6.68, 7.42 (d, J = 8.5 Hz, 4H), 7.03 (d, J = 16.30 Hz, 2H), 6.96, (d, J = 16.30 Hz, 2H), 7.42 (s, 4H); MS (FAB, m-NBA matrix): m/z 560.92 (M⁺); MS (EI): m/z 561.9, 512.9, 462.0, 436.0, 399.0, 336.0.

Page 72, line 20:

1,4-Bis(4-di-n-butylamino)styryl Anthracene: prepared from the reaction of 9,10-anthracenedicarboxaldehyde (1.17 g, 0.005 mol) and 4-dibutylaminobenzyl phosphonium iodide (7.563g, 0.0125 mol) and lithium/ethanol (0.178g, 0.0295 mol/ 70 mL). Recrystallized from N,N-dimethylformamide/water, Yield 62.4%; Anal. Calcd. for $\text{C}_{46}\text{H}_{56}\text{N}_2$: C, 86.73; H, 8.86; N, 4.39. found: C, 86.18; H, 8.59; N, 5.35; IR (KBr) ν cm^{-1} : 2948, 1607, 1518, 1365, 1186, 760; UV-Vis (acetone) λ_{max} , nm: 440, 320; ^1H NMR (C_6D_6) δ : 0.870 (t, J = 7.2 Hz, 12H), 1.18 (m, 8H), 1.48 (m, 8H), 3.08 (t, J = 7.4 Hz, 8H), 6.71, 7.51 (Ar, J = 8.5 Hz, 8H), 6.98 (d, J = 16.4 Hz, 2H), 7.82 (d, J = 16.4 Hz, 2H), 7.34, 8.66 (m, 8H, Ar); MS (FAB, m-NBA matrix): m/z 636.4 (M⁺).

Page 76, line 19:

The general procedure for the double Wittig reaction using NaOEt was followed. After 4h of reflux, the reaction mixture was cooled and water was added. Upon addition of water, an oily product formed that became a solid mass after cooling overnight. The yellow solid was obtained in 94.9% yield as a mixture of isomers. (In the above structure, the "sawtooth" bond indicates that the structure includes both cis [ans]and trans isomers.)

Page 79, line 16:

Figure 10 illustrates the reaction of 1,4-bis(cyanomethyl)benzene with 4-diphenylaminostilbene and 4-di-*n*-butylaminobenzaldehyde to yield [1,4-bis(β -cyano-4'-diphenyl-aminostyryl)benzene and 1,4-bis(β -cyano-4'-di-*n*-butylaminostyryl)benzene] 1,4-bis(β -cyano-4'-diphenylaminostyryl)benzene and 1,4-bis(β -cyano-4'-di-*n*-butylaminostyryl)benzene with donor side units and acceptors on the vinylene units (see examples 72 and 73).

Page 90, line 2:

For comparison with the data in Table 2, *para*-bis(*ortho*-methylstyryl)benzene, which lacks donor or acceptor substitution, exhibits a two-photon absorptivity of [69]55 $\times 10^{-50} \text{ cm}^4 \text{ s/photon}$ at 568 nm [C. Xu and W. W. Webb, J. Opt. Soc. Am. B, **13**, 481 (1996) and S. M. Kennedy and F. E. Lytle, Anal. Chem. **58**, 2643 (1986)]. In accord with the present invention compounds symmetrically substituted with a pair of terminal donors, [i.e., D- π -D] or symmetrically substituted to form molecules according to the donor-acceptor-donor (D-A-D) or acceptor-donor-acceptor (A-D-A) motifs exhibit substantially enhanced two-photon absorptivities compared to unsubstituted *para*-bis(*ortho*-methylstyryl)benzene.

Page 90, line 12:

Measurements of two-photon absorptivity by use of nonlinear optical transmission method in solutions of bis-donor-substituted diphenylpolyenes show a strong dependence on laser pulse duration, as indicated by the values in Table 3. These results provide evidence of nonlinear absorption that is higher-order than two-photon absorption. In particular, the results suggest that these molecules undergo two-photon induced excited state absorption, that is, two-photon absorption to an excited state and further absorption from this excited state or from a state formed rapidly on relaxation from the initial excited state. A greater excited state population is created with the longer pulses, leading to the enhanced nonlinear absorption coefficient. Additional measurements including transient photoinduced absorption measurements and picosecond time resolved pump and probe measurements confirm the presence of excited state absorption induced by two-photon absorption, which occurs at the two-photon excitation wavelength. The effective two-photon absorptivities of the series of bis-donor diphenylpolyenes is one to two orders of magnitude larger than that measured with picosecond pulses and, in accord with the present invention, these data demonstrate utility in optical limiting applications involving nanosecond laser pulses.

Furthermore, the values of the effective two-photon absorptivity obtained for the bis donor diphenylpolyenes are an order of magnitude larger than that of Rhodamine B.

Page 91, line 12:

Table 3. Effective [Two-photon]two-photon absorption data on a series of bis-donor diphenylpolyenes as measured with nanosecond (ns) and picosecond (ps) pulses and nonlinear optical transmission methods. The units of the two-photon absorptivity, δ , are $10^{-50} \text{ cm}^4\text{-s/photon}$.

Page 98, line 3:

Here we describe the utility of two-photon and multi-photon absorbing molecules as initiators of polymerization, specifically to the use of bis-donor-substituted π -conjugated molecules as two-photon initiators. Such two-photon excitable initiators have utility in two- and three-dimensional lithography applications, [for example]and in three-dimensional optical data storage with ultrahigh storage densities ($10^{12} \text{ bits/cm}^3$). When applied to a spinning disk (compact disk) type format, two-photon polymerizable materials can offer over a thousand-fold increase in storage capacity. There are two key advantages of two-photon induced polymerization relative to single-photon induced polymerization. Whereas single-photon absorption scales linearly with the intensity of the incident radiation, two-photon absorption scales quadratically. As a result, it is possible to perform two-photon polymerization with three-dimensional spatial resolution. In addition, because the phenomenon involves the simultaneous absorption of two-photons, the chromophore is excited with light of half the energy of the two-photon absorption peak. As a result, it is possible to excite molecules at a greater depth within a material than would not be possible via single-photon excitation. These two advantages also apply to for example *in vivo* photo-initiated polymerization. In lithography or stereolithography, the quadratic scaling of absorption with intensity can lead to the ability to write features below the diffraction limit of light and the ability to write features in three dimensions, which is also of interest for holography.

Page 110, line 10:

A critical issue related to [*in situ*]*in vivo* photopolymerization addressed in this invention is limitations that may be introduced by the use of visible light; specifically, visible

light does not penetrate tissues or other absorbing or scattering media at depth. To address this deficiency, we have developed photoinitiators which can be activated by two-photon absorption of near-infrared light, which has the greatest penetration depth through tissue.

Page 115, line 26:

The new multi-photon absorbers according to the present invention afford several interesting advantages for optical limiting. Firstly, these molecules exhibit effective two-photon absorptivities that are about one or more orders of magnitude higher for current state of the art materials. With such high two-photon absorptivities, these molecules are effective in limiting nanosecond duration pulses. Secondly, these molecules can have very high broadband linear transmission, since the two-photon absorption bands occur for photon energies well below the linear absorption edge. This could be a huge advantage, since low linear transmission and impairment of normal color vision are major concerns for potential users of laser protective devices. [Secondly,] Thirdly, broadband two-photon absorbing molecules have been prepared from mixtures of compounds, all of which are essentially transparent over much of the visible spectrum, each with different two-photon resonances that span a wide band. Finally, these two-photon absorbing molecules perform very well at high intensities such that good short pulse performance is assured.

Page 116, line 12:

We have demonstrated optical limiting based on the new molecular two-photon and multi-photon absorbers described herein in different optical geometries and with combinations of materials as described below. The optical limiting compositions according to the present invention may exist as crystals, mesoscopic phases, polymers, glasses, liquids or gases. The compositions may be used alone or in combination with other crystals, mesoscopic phases, polymers, glasses, liquids or gases. By "mesoscopic phases," we refer to materials with structural order on a length scale between that of individual molecules, i.e., above about 10 Angstroms, and the microscopic length scale, i.e., above about one micrometer. These materials include small molecule and polymeric liquid crystals, colloidal, micellar and liposomal suspensions, self assembled nanoparticle arrays, and the like.

Page 119, line 3:

A mechanism to enhance the two-photon absorptivity at a given wavelength is to synthesize molecules with extended conjugation such that the single-photon absorption edge

approaches the fundamental frequency of the laser, without leading to substantially decreasing linear loss. For example, simply extending the number of bridging double bonds in the stilbene derivatives from one to five double bonds enhances the effective two-photon absorptivity by a factor of five. The enhanced two-photon absorption is evidenced in Figure 19 where the nonlinear transmission of 4,4'-bis-(di-n-butylamino)stilbene is compared with [1-(di-4-n-butylaminophenyl)-10-(4-dimethylaminophenyl) deca 1, 3, 5, 7, 9-pentaene] 1-(di-4-n-butylaminophenyl)-10-(4-dimethylaminophenyl) deca-1, 3, 5, 7, 9-pentane.